SOME CONTRIBUTIONS TO DESIGN OF EXPERIMENTS USING F-SQUARES

THESIS

SUBMITTED FOR THE AWARD OF THE DEGREE OF

Doctor of Philosophy

In

Statistics

By

SANGHMITRA SHARMA

Under the Supervision of

Dr. BUSHRA HUSAIN

DEPARTMENT OF STATISTICS AND OPERATIONS RESEARCH
ALIGARH MUSLIM UNIVERSITY
ALIGARH (INDIA)

2016

Dedicated

to

My Mother-in-Law

Late. Mrs. Shanti Sharma

Department of Statistics & Operations Research
Aligarh Muslim University
Aligarh-202002, (India)

Candidate's Declaration

I, **Sanghmitra Sharma**, Department of **Statistics & Operations Research** certify that the work embodied in this Ph.D. thesis is my own bonafide work carried out by me under the supervision of **Dr. Bushra Husain** at Aligarh Muslim University, Aligarh. The matter embodied in this Ph.D. thesis has not been submitted for the award of any other degree.

I declare that I have faithfully acknowledged, given credit to and referred to the research workers wherever their works have been cited in the text and the body of the thesis. I further certify that I have not willfully lifted up someone else's work, paragraph, text, results, etc. reported in the journals, books, magazines, reports, dissertations, theses, etc., or available at web-sites and included them in this Ph.D. thesis and cited as my own work.

Date: *(Signature of the candidate)*

Certificate from the Supervisor

This is to certify that the above statement made by the candidate is correct to the best of my knowledge.

(Dr. Bushra Husain)
Assistant Professor

(Signature of the Chairman of the Department with seal)

Department of Statistics & Operations Research
Aligarh Muslim University
Aligarh-202002, (India)

Course Work/Comprehensive Examination/Pre-Submission Seminar Completion Certificate

*This is to certify that **Mrs. Sanghmitra Sharma**, Department of **Statistics & Operations Research** has satisfactorily completed the course work/comprehensive examination and pre-submission seminar requirement, which is part of her Ph.D. (Statistics) programme.*

Date: ***(Signature of the Chairman of the Department)***

Department of Statistics & Operations Research
Aligarh Muslim University
Aligarh-202002, (India)

Copyright Transfer Certificate

Title of the Thesis	**:**	**Some Contributions to Design of Experiments Using F-Squares**
Candidate's Name	**:**	**Sanghmitra Sharma**

Copyright Transfer

The undersigned hereby assigns to the Aligarh Muslim University, Aligarh copyright that may exist in and for the above thesis submitted for the award of the Ph.D. degree.

(Signature of the candidate)

Table of Contents

ACKNOWLEDGEMENT

First and foremost, I praise God, the Almighty, providing me this opportunity and granting me the capability to complete this research work successfully. This thesis appears in its current form due to the assistance and guidance of several people during this hard time, but a very fruitful period of new experiences. I would therefore like to offer my sincere thanks to all of them.

I feel great pleasure at the completion of this thesis entitled ***"Some contributions to design of experiments using F-squares"*** *and would like to show deep indebtness and sincere gratitude to all who helped me in the completion of this work. Undertaking this Ph.D has been a truly life changing experience for me and it would not have been possible to do without the support and guidance. I received from many people.*

I would like to express my indebtness and sincere gratitude to my supervisor ***Dr. Bushra Husain****, Department of Statistics and Operations Research, Women's College, Aligarh Muslim University, Aligarh, for her valuable suggestions, continued interest, encouragement and support throughout the completion of this work. I highly appreciate the way she encouraged me to work hard as well as the way she refined my aptitude for research. Her excellent guidance, advice towards the structure of this thesis is very much appreciated. I have always felt privileged and honoured to work under her guidance. It has always been a pleasure working with her. To be honest I am extremely impressed by her personality and teaching methods. I would like to thank her for giving me the opportunity to do research.*

I am thankful to ***Prof. Q. M. Ali,*** *Chairman, Department of Statistics and Operations Research, Aligarh Muslim University, Aligarh, for his cooperation, encouragement and furnishing all necessary facilities to complete this work in time.*

*I would like to thank my teachers **Prof. Ariful Islam,** my M.Phil. Supervisor, **Dr. Athar Ali Khan, Dr. S. S. Hasan, Dr. R. U. Khan, Dr. Haseeb Athar** and **Dr. Mohd. Arshad** for their concern and consultancy.*

I would also like to thank all the faculty and non-teaching staff members in the department of Statistics and Operations Research, A.M.U., Aligarh, for their help during the course of my study here.

*I am greatly indebted to my research colleagues and friends who have encouraged me throughout this work. I am particularly thankful to **Dr. Neha Gupta, Dr. Zubdah-e-Noor, Dr. Sana Iftekhar, Mrs. Anamika Kulshrestha, Dr. Shamsher Khan, Mrs. Sadiya Parveen** and **Ms. Afrah** among others for their cooperation and sharing of ideas during the preparation of this work.*

*Finally, I would like to express my deep sense of respect and thanks to my loving parents **Mrs. Laxmi Sharma** and **Mr. Ramesh Chandra Sharma** for their love, prayers and care. I am indebted to my mother-in-law (**Late.) Mrs. Shanti Sharma**. I take the opportunity to thank my husband **Mr. Manish Dutt Sharma**, but I have no words in my command that can express my gratitude towards him for his moral support and prayers which have enabled me to reach this stage. He has seen very hard times day in and day out and sacrificed the joys of life for educating and preparing me for this goal of my life. I am extremely thankful to my son (**Ojasva Sharma**) and daughter (**Dakshita Sharma**) and all my relatives for their continuous support, endless patience, blessings and moral support.*

I am also thankful to University Grants Commission, New Delhi for providing me the fellowship during my research work.

Last but not the least; I thank each and every one who has helped me directly or indirectly at some stage or the other in making this thesis possible. Thanks!

Date: …………… **Sanghmitra Sharma**

Place: Aligarh *sanghmitra2007@rediffmail.com*

PREFACE

This thesis entitled "**Some Contributions to Design of Experiments using F-Squares**" is submitted to the Faculty of Science, Aligarh Muslim University, as a partial fulfillment of the requirements to obtain the Ph.D. degree in Statistics. It embodies the research work carried out by me in the Department of Statistics and Operations Research, Aligarh Muslim University, Aligarh.

In Mixture Experiments, the response of interest depends only on the proportions of the components or ingredients of the mixture, but does not depend on the quantity of the components. The empirical models used to describe the characteristics of these products with the proportions of their ingredients are called *mixture models* and the related designs of experiments are called *mixture designs* or *simplex designs*. Fifty-eight years ago, Scheffé laid the foundation for the development of mixture tools (designs and models) by introducing the simplex lattice designs and their associated canonical polynomials. If an individual component proportion x_i is unity then the mixture comprises of a single ingredient and is called a "*single-component*" mixture or a *pure* mixture. Each run must satisfy

$$x_1 + x_2 + \cdots + x_q = 1, \text{ with } 0 \le x_i \le 1, \text{ for } i = 1, 2, \ldots, q.$$

For example, lemonade is made by mixing lemon juice, salt, sugar and black salt in different proportions and the response of interest is the tangy flavour of the lemonade. This is a five component mixture experiments where the fifth component is water.

Main contributors for experiments with mixtures are Scheffé (1958; 1963), Gorman and Hinman (1962), Kurotori (1966), McLean and Anderson (1966), Murthy and Das (1968), Thompson and Myers (1968), Saxena and Nigam (1973), Cornell (1973, 1975, 1995 and 2000), Snee (1973, 1975 and 1979), Hare (1974; 1979), Snee and

Marquardt (1974; 1976), John (1984), Czitrom (1988), Hilgers and Heiligers (2003), amongst others.

For obtaining blocked mixture experiments, we ensure that the groups of mixture blends differ from the other groups or blocks by an additive constant. Further for orthogonal blocking of the mixture blends, we require the estimates of the blending properties in the fitted model to be uncorrelated with the block effects. In mixture experiments, we may run one or more of the blocks at each combination of the process variables. John (1984) gave the following conditions for the orthogonal blocking of blends for Scheffé's quadratic model when the blocks are of the same size.

$$\sum_{u=1}^{n_w} x_{iu} = k_{1w} \qquad \text{(constant);} \qquad i = 1, 2, \ldots, q$$

$$\sum_{u=1}^{n_w} x_{iu} x_{ju} = k_{2w} \qquad \text{(constant);} \qquad 1 \leq i < j \leq q$$

where $w = 1, 2, \ldots, t\text{-}1$ with t as the number of blocks.

Main contributors for orthogonally blocked experimental designs are Kiefer (1959, 1975 and 1978), Nigam (1970, 1976), John (1984), Laywine (1989), Czitrom (1988, 1989, 1992), Draper et al. (1993), Prescott et al. (1993), Lewis et al. (1994), Prescott et al. (1997), Chan and Sandhu (1999), Ghosh and Liu (1999), Aggarwal et al. (2002) and Singh (2003).

F-squares and orthogonal F-squares are a generalization of latin squares and orthogonal latin squares. Hedayat and Seiden (1970) gave the following definition of an F-square:

***Definition* :** Let $\mathbf{A} = [a_{ij}]$ be an $n \times n$ matrix and let $\mathbf{\Sigma} = (c_1, c_2, \ldots, c_m)$ be the ordered set of distinct elements of $\mathbf{A}$. In addition, suppose that for each $k = 1, 2, \ldots, m$, c_k appears precisely λ_k times ($\lambda_k \geq 1$) in each row and in each column of $\mathbf{A}$. Then, $\mathbf{A}$ is called a frequency square or more concisely, an F-square of order n on $\mathbf{\Sigma}$ with frequency vector $(\lambda_1, \lambda_2, \ldots, \lambda_m)$ and is denoted by $F(n; \lambda_1, \lambda_2, \ldots, \lambda_m)$.

Main contributors for F-squares are Finney (1945, 1946a, 1946b), Freeman (1966), Addleman (1967), Hedayat and Seiden (1970) and Hedayat et al. (1975). F-square based orthogonally blocked mixture experiments have been considered by Aggrawal et al. (2008, 2009, 2011a, 2011b and 2013).

The uniform design (UD) is one kind of space filling design and is widely used in computer experiments. Let n denote the number of runs, s the number of input variables and T the input space in a system where one wants to implement computer experiments. A uniform design seeks n points, denoted by $\mathcal{P}_n = \{x_1, \dots, x_n\}$, to be uniformly scattered on T. For given (n, s, T), we require a measure of uniformity and a way of finding a uniform design.

Some commonly used measures of uniformity are star discrepancy, symmetrical discrepancy, centered L_2-discrepancy and Wrap-around discrepancy. There is a unique UD $\left\{\frac{1}{2n}, \frac{3}{2n}, \dots, \frac{(2n-1)}{2n}\right\}$ for the case of one input variable (i.e., s = 1) under the star discrepancy, the symmetrical discrepancy, the centered L_2-discrepancy and Wrap-around discrepancy.

Main contributors for uniform designs and measures of discrepancies are Warnock (1972), McKay et al. (1979), Fang (1980), Fang and Wang (1981), Niederreiter (1978, 1992), Shaw (1988), Bundschuh and Zhu (1993), Wang and Fang (1996), Fang and Li (1995), Ma (1997a; 1997b), Fang and Winker (1998), Hickernell (1998a, 1998b), Fang et al. (1999), Hickernell et al. (2000), Fang et al. (2000) and Fang et al. (2001), amongst others.

THESIS OUTLINE

The research work presented in this thesis is based on contributions to Design of Experiments using F-squares for optimal experimental designs and Uniform Designs. This thesis is spread over five chapters. A comprehensive bibliography has also been given at the end, which has been referred to during the research work.

CHAPTER 1 is an expository in mixture designs and provides a brief review of the ideas about the general mixture problems for understanding the different concepts as regards mixture experiments and uniform designs with low discrepancy.

CHAPTER 2 is focused on F-square based D-, A-, and E-optimal orthogonally blocked designs in four mixture components for inverse models.

CHAPTER 3 presents D-, A- and E-optimal designs and optimal orthogonally blocked designs based on F-squares for reduced cubic canonical model in four mixture components.

CHAPTER 4 presents Uniform Designs based on cyclic F-squares with low discrepancy (L_2-discrepancy, Centered L_2-discrepancy and Wrap-around discrepancy).

CHAPTER 5 deals with Centered L_2-discrepancy and D-, A- and G-efficient uniform designs for mixture experiments in three and four components based on F-squares.

Chapter 1

BASIC CONCEPTS OF MIXTURE THEORY

1.1. Introduction

Mixtures are everywhere. Any two or more items may be combined together to form mixtures. In the usual mixture experiments, the proportions of the mixture ingredients influence the response of interest entirely. Consider the following examples (Cornell (2002)) of the products where two or more ingredients are combined in order to obtain an end product.

a. The driving speed and automobile size may affect the blending behaviours of fuels being tested to compare the average mileage of the fuels individually as well as when blended together.
b. Cake baking formulations using flour, sugar, condensed milk and baking powder.
c. Building construction concrete formed by mixing sand, water and one or more types of cement.
d. Fruit punch consisting of juices from watermelon, pineapple and orange.
e. Making sandwich patties using flour, salt and cheese.

Usually in a mixture experiments, the response of interest depends only on the proportions of the components or ingredients of the mixture, but does not depend on the quantity of the components. If the property of interest depends only on the relative proportions (or percentages) of the ingredients present in the mixture and is independent of the total amount of the mixture, then the empirical models used to describe the characteristics of these products with the proportions of their ingredients are called *mixture models* and the related designs of experiments are called *mixture designs* or *simplex designs*. In the examples cited above, the properties of interest are as follows:

a. The mileage and controlling power obtained after mixing the fuels.
b. The fluffiness and layer appearance of cake.
c. The hardness and compression strength of the concrete.
d. The fruitiness flavour of the punch.
e. The texture and flavour of patties.

Fifty-eight years ago, Scheffé's (1958) pioneering article "*Experiments with Mixtures*", laid the foundation for the development of mixture tools (design and models) by introducing the simplex lattice designs and their associated canonical polynomials.

If x_i (i = 1, 2, …, q) denotes the proportion of the i^{th} ingredient in the mixture then the *mixture constraints* for each run must satisfy

$$x_1 + x_2 + \cdots + x_q = 1, \text{ with } 0 \leq x_i \leq 1, \text{ for } i = 1, 2, \ldots, q. \tag{1.1}$$

In the lemonade example, let the lemon juice comprise 75% of the lemonade (or x_1 = 0.75), salt 5% of the lemonade (or x_2 = 0.05), sugar 15% of the lemonade (or x_3 = 0.15) and black salt 5% of the lemonade (or x_4 = 0.05). If in (1.1) an individual proportion x_i is unity then the mixture comprises of a single ingredient and is called a "*single-component*" mixture or a *pure* mixture.

Due to the constraints (1.1) on x_i, the geometric description of the factor space containing the q-components consists of all points on or inside the boundaries (vertices, edges, faces, etc.) of a regular (q - 1) dimensional simplex. The simplex factor space is a straight line for two components. With three components q = 3, the simplex factor space is an equilateral triangle. The coordinate system used for the values x_i, i = 1, 2, ..., q is called a *simplex coordinate system*.

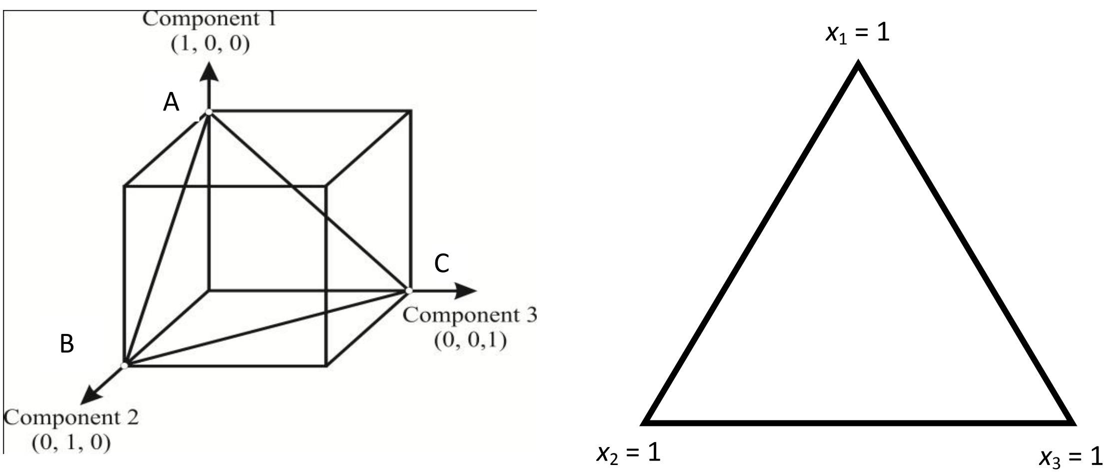


Figure 1.1. Three-component simplex region

ΔABC is the three-component triangle where $x_i \geq 0$ and all experimental points must lie on or inside the triangle satisfying the equation $x_1 + x_2 + x_3 = 1$. For four components, the simplex is a tetrahedron as shown in Figure 1.2.

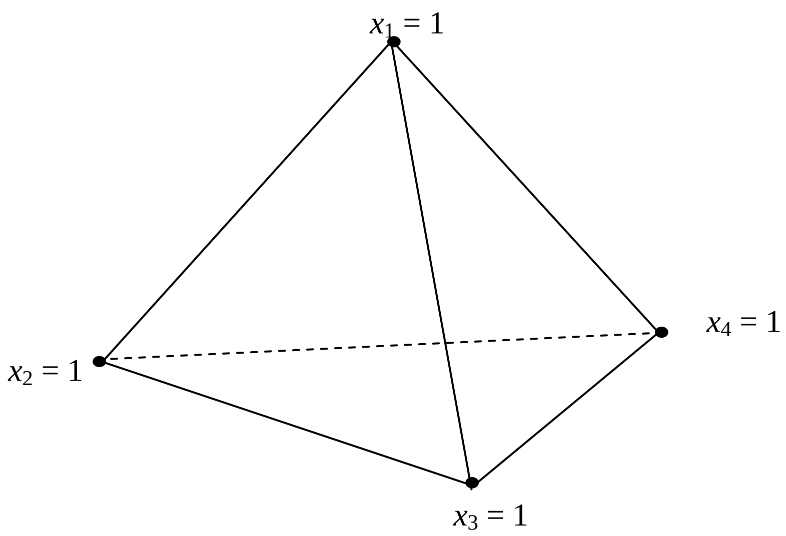


Figure 1.2. Four-component simplex region

The coordinate system used for the values x_i, $i = 1, 2, \ldots, q$ is called a simplex coordinate system. In Figure 1.1, we see the vertices of the simplex or ΔABC represent the single-component mixtures and are denoted by $x_i = 1$, $x_j = 0$ for $i, j = 1, 2$ and 3, $i \neq j$. The interior points of the triangle represent mixtures in which none of the three components is absent; that is, $x_1 > 0$, $x_2 > 0$ and $x_3 > 0$. The centroid of the triangle corresponds to the mixture with equal proportions (1/3, 1/3, 1/3) from each of the components. Figure 1.2 represents the tetrahedron in four components having proportions x_1, x_2, x_3 and x_4.

1.2. Simplex - Lattice Design

A $\{q, m\}$ simplex lattice design for q factors (components) is defined by all possible combinations of component levels with the proportions being

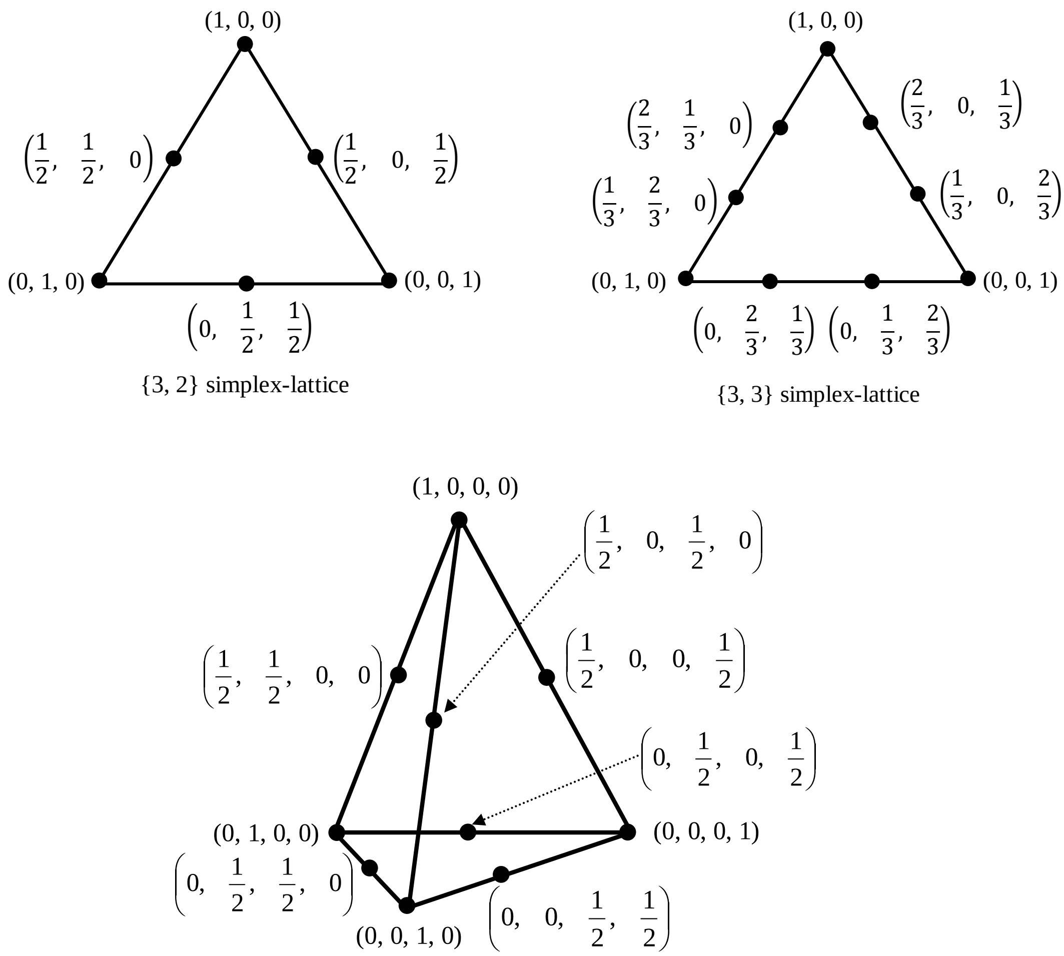
$$x_i = 0, \frac{1}{m}, \frac{2}{m}, \ldots, 1, \; i = 1, 2, \ldots, q \qquad (1.2)$$

As an example, the {3, 3} simplex lattice design consists as factor levels all the possible blends of the three components with the proportions satisfying (1.2). The {3, 3} simplex lattice consists of the nine points on the boundary of the triangular factor space.

The three vertices (1, 0, 0), (0, 1, 0) and (0, 0, 1) represent the individual components whereas the other six points represent ternary blends or three-component mixtures, with each component contributing 1/3 and 2/3 of the blend. The {3, 2}, {3, 3} and {4, 2} simplex- lattice are shown in Figure 1.3.

Table 1.1 Simplex Lattice Design

x_1	x_2	x_3
1	0	0
0	1	0
0	0	1
2/3	1/3	0
1/3	2/3	0
0	2/3	1/3
0	1/3	2/3
1/3	0	2/3
2/3	0	1/3



{4, 2} simplex -lattice

Figure 1.3. The {3, 2}, {3, 3} and {4, 2} simplex-lattice arrangements and the coordinates setting of the design points.

The number of design points in the $\{q, m\}$ simplex-lattice is $(q + m - 1)! / (m!(q - 1)!)$. Scheffé (1958) defined canonical polynomials to support his simplex-lattice designs by modifying the usual models in x_i using the simplex restriction $x_1 + x_2 + \cdots + x_q = 1$. In particular, let us consider two components whose proportions are represented by x_1 and x_2, with $x_1 + x_2 = 1$. The first-degree polynomial having two component proportions as x_1 and x_2 is as follows

$\eta = \beta_0 + \beta_1 x_1 + \beta_2 x_2$, where η represents the expected response and

$\beta_0 = \beta_0\,(x_1 + x_2)$

then we get

$$\eta = (\beta_0 + \beta_1)\,x_1 + (\beta_0 + \beta_2)\,x_2$$

$= \beta_1' x_1 + \beta_2' x_2$, so that the constant term β_0 is removed from the model.

In general, the canonical forms of the mixture models are as follows:

Linear:
$$\eta = \sum_{i=1}^{q} \beta_i x_i \tag{1.3}$$

Quadratic:
$$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum^{q} \beta_{ij} x_i x_j \tag{1.4}$$

Special cubic:
$$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum^{q} \beta_{ij} x_i x_j + \sum\sum_{i<j<k}\sum^{q} \beta_{ijk} x_i x_j x_k \tag{1.5}$$

Full cubic:
$$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum^{q} \beta_{ij} x_i x_j + \sum_{i<j}\sum^{q} \gamma_{ij} x_i x_j (x_i - x_j) + \ldots + \sum\sum_{i<j<k}\sum^{q} \beta_{ijk} x_i x_j x_k . \tag{1.6}$$

The number of terms in the $\{q, m\}$ polynomials is a function of m, the degree of the equation, as well as the number of components q. The numbers of terms for several values of q are listed in Table 1.2.

Table 1.2. Number of terms in the canonical polynomials

Number of components q	Linear	Quadratic	Special Cubic	Full Cubic
2	2	3	-	-
3	3	6	7	10
4	4	10	14	20
5	5	15	25	35
6	6	21	41	56
7	7	28	63	84
8	8	36	92	120
⋮	⋮	⋮	⋮	⋮
q	q	$q(q+1)/2$	$q(q^2+5)/6$	$q(q+1)(q+2)/6$

The terms in the canonical mixture polynomials have simple interpretations. In the linear and quadratic forms (1.3) and (1.4), if $x_i = 1$, then $\eta = \beta_i$. The parameter β_i represents the expected response to the pure mixture and pure component i and the above polynomials given by $\sum_{i=1}^{q} \beta_i x_i$ is called the linear blending proportion. The number of terms for the canonical forms of the mixture models (1.3), (1.4), (1.5) and (1.6) are as shown in Table 1.2.

1.3. Simplex - Centroid Design

Another type of mixture design is simplex-centroid design. Scheffé (1963) introduced the simplex centroid designs. It is an alternative to the $\{q, m\}$ simplex-lattice designs. In a q-component simplex-centroid design, the number of distinct points is $2^q - 1$. These points correspond to the q permutations of (1, 0, …, 0) or q single component blends, the $\binom{q}{2}$ permutations of (1/2, 1/2, 0, …, 0) or all binary mixtures, the $\binom{q}{3}$ permutations of (1/3, 1/3, 1/3, 0, …, 0), …, and so on, with the overall centroid point ($1/q$, $1/q$, …, $1/q$) or q-nary mixture.

Scheffé (1963) defined the special canonical polynomial in q components for the simplex centroid design as

$$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum^{q} \beta_{ij} x_i x_j + \sum_{i<j<k}\sum\sum^{q} \beta_{ijk} x_i x_j x_k + \ldots + \beta_{1,2,\ldots,q} x_1 x_2 \ldots x_q. \quad (1.7)$$

The parameter β_i represents the expected response of the pure mixture x_i and is called the linear blending value of component x_i. The parameter β_{ij} represents the coefficient of the nonadditive blending of component x_i and x_j. The other parameters in the polynomial may be defined similarly. The $2^q - 1$ parameters in the polynomial in equation (1.7) equals the number of points in the simplex centroid designs.

For example, four component simplex centroid design consists of $2^4 - 1 = 15$ points which are the four vertices, the mid points of the six edges, the centroids of the four faces, and the simplex-centroid design points are (1, 0, 0, 0), (0, 1, 0, 0), (0, 0, 1, 0), (0, 0, 0 ,1), (1/2, 1/2, 0, 0), (1/2 ,0 , 1/2, 0), …, (0, 0, 1/2, 1/2), (1/3, 1/3, 1/3, 0), …, (0, 1/3, 1/3, 1/3), (1/4, 1/4, 1/4, 1/4). Thus, the simplex centroid designs are better space filling designs as compared to the $\{q, 2\}$ simplex-lattices.

The arrangement of the points of the simplex centroid design for three and four components are shown in figures 1.4 (a) and 1.4 (b), respectively.

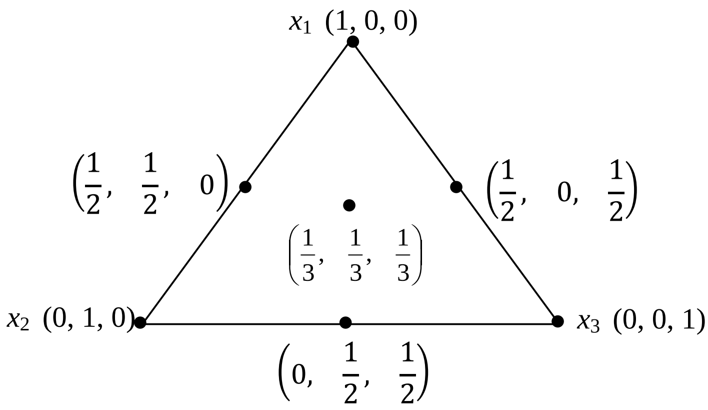


Figure 1.4(a). Simplex centroid design for three components.

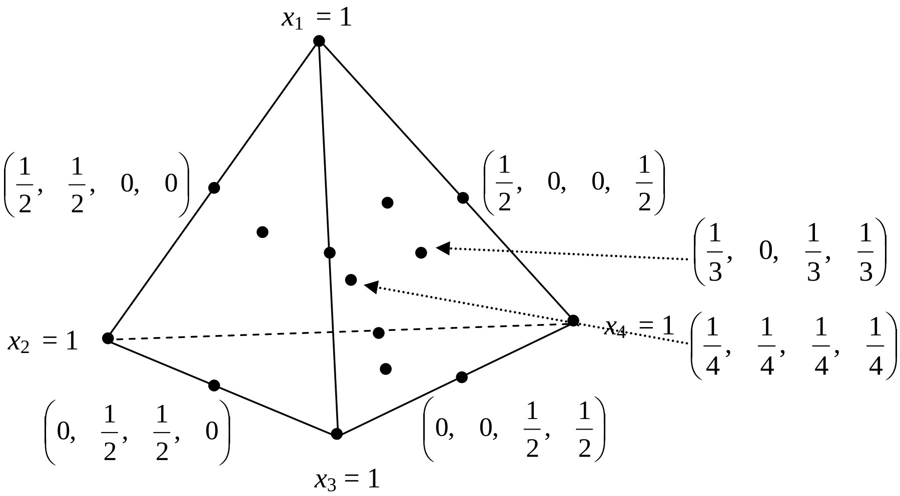


Figure 1.4(b). Simplex centroid design for four components.

Murthy and Das (1968) considered the generalized forms of Scheffé's designs and explored the entire factor space uniformly by developing the symmetric-simplex designs. Saxena and Nigam (1973) presented the symmetric-simplex block designs for experiments with mixtures.

1.4. Axial Design

The points of the $\{q, m\}$ simplex-lattice and q - component simplex-centroid designs are positioned on the boundaries (vertices, edges, faces, etc.) of the simplex factor space. Axial designs comprise mainly of complete mixture or q - component blends where most of the points are positioned inside the simplex (Cornell (1975)).

The axis of component i is the imaginary line extending from the base points $x_i = 0$, $x_j = 1 / (q-1)$ for all $i \neq j$ to the vertex where $x_i = 1$, $x_j = 0$ for all $i \neq j$.

An *axial* design's points are positioned only on the components axes. The simplest form of axial design is the one whose points are positioned equidistant from the centroid $(1/q, 1/q, \ldots, 1/q)$ towards each of the vertices. x_i is the unit of the measuring distance from centroid that is denoted by Δ and the maximum value for Δ is $(q - 1)/q$. The concept of axial design was suggested by Cornell (1975).

If the effects of the components are to be measured or if the first-degree models are to be fitted in screening experiments, then we may use axial design. Snee and Marquardt (1974) and Chan et al. (1998) studied D-optimal axial designs for quadratic and cubic additive mixture models which were introduced by Darroch and Waller (1985) and compared the saturated D-optimal axial design and D-optimal design for the quadratic model in terms of their efficiency and uniformity. Figure 1.5 depicts a three component axial design where the distance from the center of the simplex to the points is Δ.

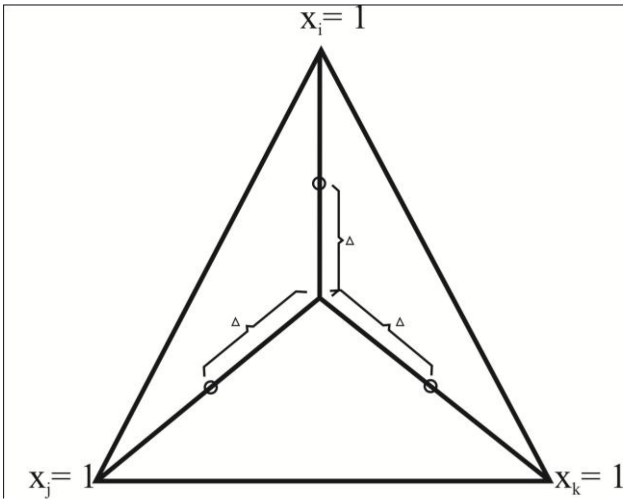


Figure 1.5. A three-component axial design where the distance from the center of the simplex to the points is Δ.

1.5. Constraints on the Mixture Components

In mixture experiments, one is not completely at freedom to explore the entire simplex region because of certain additional limitations when there are constraints on the component proportions. The constrained mixture problem has the following general form in the presence of both the upper and/or lower bound constraints of the form $L_i \le x_i \le U_i$, $i = 1, 2, \ldots, q$, where L_i is the lower bound for the i^{th} component and U_i is the upper bound for the i^{th} component.

$$x_1 + x_2 + \cdots + x_q = 1$$
$$L_i \le x_i \le U_i, \text{ for } i = 1, 2, \ldots, q$$
$$\text{with } L_i \ge 0 \text{ and } U_i \le 1$$

The "pseudo" components are defined as the combination of original components and the primary reason for introducing the pseudocomponents is that usually both the construction of designs and model fitting becomes comparatively easier. The experimental region is a simplex in the presence of upper bounds alone, if and only if it lies entirely within the original simplex. Generally, the shape of the polyhedron is highly complicated as compared to that of the simplex because the polyhedron has more than q vertices and more than q edges. This makes construction of the design and the fitting of the model easier as compared to the constrained region of interest. The lower bound constraints are expressed in general form

$$0 \le L_i \le x_i, \qquad \text{for } i = 1, 2, \ldots, q$$

The new component x_i' called L-pseudocomponents were defined by Kurotori (1966) as

$$x_i' = \frac{x_i - L_i}{1 - L}$$

where,

$$L = \sum_{i=1}^{q} L_i < 1 \qquad \text{denotes the sum of all the lower bounds.}$$

Crosier (1984) suggested defining the U–pseudocomponents. Cornell (2002) denoted the U–pseudocomponents by u_i using the following formula

$$u_i = \frac{U_i - x_i}{U - 1} \qquad i = 1, 2, \ldots, q$$

where

$$U = \sum_{i=1}^{q} U_i > 1 \qquad \text{denotes the sum of all the upper bounds.}$$

The experimental region is a simplex in the presence of upper bounds alone, if and only if it lies entirely within the original simplex. The shape of the polyhedron is generally highly complicated as compared to the simplex because the polyhedron has more than q vertices and more than q edges. The lower and upper bound constraints are checked for their consistency in order to determine the coordinates of the extreme vertices of the constrained region. If one or more of the bounds cannot be attained, then the constraints are adjusted to make them consistent and then a formula is used to determine the number of extreme vertices, edges and two-dimensional faces of the constrained region.

Scheffé (1958) considered the case where the components are restricted below by certain bounds. Scheffé (1958) suggested modification of the $\{q, m\}$ lattice by introducing pseudocomponents for each of the restricted components. For the sake of setting up designs and fitting models in a simpler form when the component proportions are restricted by bounds, Kurotori (1966) suggested L-pseudocomponent transformation when only lower bound restrictions are imposed on x_i. When the sub region of the simplex is also a simplex then the use of L-pseudocomponent simplifies the construction of designs by allowing the $\{q, m\}$ simplex-lattice or simplex-centroid designs to be used in the L-pseudocomponent system. Gorman (1970) used the inverse transformation from the pseudocomponents back to the original components in order to yield a fitted model in the original components. Saxena and Nigam (1977) suggested a judicious choice of the symmetric-simplex design in order to explore the interior of a highly constrained region. The consistent constraint regions in mixture experiments were presented by Piepel (1983a). The centroids in constrained mixture experiments can be calculated by using the procedure described in Piepel (1983b). Crosier (1984) defined U-pseudocomponents when only upper bound constraints are imposed on the mixture component proportions. Crosier (1986) gave formulas for enumerating the number of extreme vertices, edges and two-dimensional faces of the constrained region. McLean and Anderson (1966) introduced the Extreme Vertices Design (EVD) for this situation, which generates design points from the extreme vertices, edge centroids and face centroids of the constrained region. For designs with many components, the number of extreme vertices can greatly outnumber the terms in the Scheffé models. Snee and Marquardt (1974) introduced the XVERT algorithm, which can select a subset of design points from a candidate list consisting of extreme vertices and centroids. McLean and Anderson's (1966) extreme Vertices (EV) algorithm, Snee and Marquardt's (1974) XVERT algorithm and Nigam et al.'s (1983) XVERT1 algorithm are some of the procedures for calculating the coordinates of the extreme vertices of a constrained region.

Prescott and Draper (1998) considered the case when the experimenter is not in a position to explore the entire simplex due to the additional constraints L_i and/or U_i imposed on some or all of the x_i's in the mixture for Scheffé's quadratic model. For such cases, Prescott and Draper (1998) obtained D-optimal orthogonal block designs and demonstrated that the restricted region may be simplified by the use of the pseudo components by developing designs for the particular case where $L_i = 0$ for i = 1, 2, ..., q-1. D-optimal designs in two orthogonal blocks for Darroch and Waller's quadratic model in constrained mixture components were obtained by Aggarwal and Singh (2006).

1.6. Other Mixture Model Forms

Scheffé's polynomial models are adequate for well-behaved systems. For other types of systems, functional forms other than the Scheffé polynomials are available in the literature. Draper and John (1977) proposed mixture models with inverse terms in order to model extreme changes in the response when the value of one or more component tends to a boundary of a simplex region (i.e., where one or more $x_i \rightarrow 0$). Sometimes in mixture experiment investigations, it is worthwhile to study the relationship in the form of ratios of one or more of the components to the other components in the mixtures. Standard polynomial models may be written in the ratio-variables by defining q-1 ratios as variables. The models obtained may also be fitted to data collected at the points of standard factorial arrangements. Cox's (1971) models allow for a different interpretation of the coefficients from Scheffé's models, but give the same predicted responses. Cox (1971) presented a note on polynomial response functions for mixtures where the parameters express the relative changes in the response by comparing the value of response in the simplex against the value of response in the mixtures. For a comparative type of experiment, the interpretations of the parameter estimates are closer in meaning to the treatment effects in the presence of certain restrictions imposed on the parameter values in the standard polynomial models. Smith and Beverly (1997) generated the linear and quadratic Cox mixture models with useful properties. Aitchison and Bacon-Shone (1984) introduced the standard polynomial models which depend on the component proportions and involve contrasts between pairs of log x_i where i = 1, 2, …, q. The inactivity of the components as well the additive effects of the components may be tested by using the log contrast models. Piepel and Cornell (1994) and Cornell (2000) studied the slack-variable models.

1.6.1 Becker's Model

For the situations when some components have additive effects, Becker (1968) introduced the following three mixture models.

$$\eta = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j} \beta_{ij} f(x_i,\ x_j) + \ldots + \sum_{i_1<i_2<\ldots<i_n} \beta_{i_1 i_2 \ldots i_n} f(x_{i_1},\ x_{i_2},\ \ldots,\ x_{i_n}), \tag{1.8}$$

where

$$f(x_{i_1}, x_{i_2},\ \ldots, x_{i_n}) = \min(x_{i_1},\ x_{i_2},\ \ldots,\ x_{i_n}) \qquad \text{for Model } H_1$$

$$= \frac{(x_{i_1} x_{i_2} \ldots x_{i_n})}{(x_{i_1} + x_{i_2} + \ldots + x_{i_n})^{n-1}} \qquad \text{for Model } H_2$$

$$= (x_{i_1} x_{i_2} \ldots\ x_{i_n})^{1/n} \qquad \text{for Model } H_3$$

and $2 \le n \le q$. If in model H_2 any denominator is zero, the value of corresponding term is taken as zero. The models are applied in different scientific areas and are homogenous of degree one.

Becker (1968) fitted the above models to the Lee and Warner's (1935) freezing points data of the ternary system biphenyl (B_1)-bi benzyl (B_2)-naphthalene (B_3) and found that model H_1 performs better than any of the third order models. He concluded further that the additive properties of the models H_2 and H_3 make them superior to the polynomial models. However, the non-centrality of the response surface is dealt more efficiently by the polynomial models. Snee (1973) cited the flare experimental data as an example where Becker's minimum model gives a better fit than the polynomial models. Hilgers (2000), Hilgers and Heiligers (2003) and Cornell (2002) have described various situations where models (1.8) were applied and found to be a better fit than the polynomial models. Aggarwal et al. (2013) obtained D-, A- and E-optimal orthogonal block designs for four mixture components in two blocks for Becker's models and K-model.

1.6.2 Model with Inverse Terms

Draper and St. John (1977) presented the following mixture models with inverse terms for modeling extreme changes in the response as the value of certain components tends to the boundary (usually the 'zero' boundary)

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{-i} x_i^{-1}$$

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q} \beta_{-i} x_i^{-1}$$

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q}^{q} \beta_{ij} x_i x_j + \sum_{1 \le i < j < k \le q}^{q} \beta_{ij} x_i x_j x_k + \sum_{i=1}^{q} \beta_{-i} x_i^{-1}$$

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q}^{q} \beta_{ij} x_i x_j + \sum_{1 \le i < j < k \le q}^{q} \beta_{ij} x_i x_j x_k + \sum_{1 \le i < j \le q}^{q} \gamma_{ij} x_i x_j (x_i - x_j) + \sum_{i=1}^{q} \beta_{-i} x_i^{-1} \tag{1.9}$$

The model is an augmentation of the Scheffé's polynomials with the additional terms of the form x_i^{-1} added in order to address the possible edge effects (Draper and St. John (1977)) in the response as x_i approaches zero boundary of the simplex. It is assumed that the value of x_i never reaches zero, but that the value could be very close to zero; i.e. $x_i \rightarrow \varepsilon_i > 0$; where ε_i is a very small positive quantity. Some prior work involves the use of inverse terms in x_i applied to crop yields by Nelder (1966) and applications to data from plant nutrition experiments by Clarke (1968) and Clarke and Esan (1971).

1.6.3 Darroch and Waller's Model

Darroch and Waller (1985) suggested the following additive polynomial model for experiments with mixtures

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{ii} x_i^2 \tag{1.10}$$

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \sum_{j=1}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q} \beta_{ii} x_i^3 \tag{1.11}$$

The terms of degree more than one in (1.10) are pure quadratic terms and in (1.11) are pure quadratic and cubic terms added to Scheffé's linear blending model. This model is additive in the mixture components in the sense that it is a sum of separate functions of x_1, x_2, …, x_q. When mixture components x_1, x_2, …, x_q differ but the sums $x_1 + x_2 + \cdots + x_s$ and $x_{s+1} + \cdots + x_q$ are fixed, $(1 \le s < q)$ then the total effect is the summation of the effects of varying $x_1 + x_2 + \cdots + x_s$ and $x_{s+1} + \cdots + x_q$ separately. When mixture components have additive effects on the response function, then we may use this model for the design of industrial products.

Chan et al. (1998a) introduced A-optimal weighted simplex-centroid designs for Darroch and Waller's quadratic polynomial model while Chan et al. (1998b) obtained D-optimal saturated axial designs for quadratic and cubic additive mixture models. Aggarwal et al. (2008) studied orthogonal blocking of blends for Darroch and Waller's quadratic model using F-squares in some components which assume equal volume fraction and have also given the D-, A- and E-optimalities of the different designs for $q = 4$.

1.6.4 K-Model

Draper and Pukelshiem (1997) proposed a set of mixture models referred to as K-models. K-models offer alternative depiction of the mixture models. These are based on the kronecker algebra of vectors and matrices. K-models have attractive symmetries and compact notation and are homogeneous model functions. Many advantages of the Kronecker model viz; the homogeneity of regression terms and reduced ill-conditioning of information matrices have been listed by Draper and Pukelshiem (1999) and Prescott et al. (2002).

$$E(y) = (x \otimes x)\theta = \sum_{1 \le i, j \le q} \theta_{ij} x_i x_j = \sum_{i=1}^{q} \beta_{ii} x_i^2 + \sum_{1 \le i < j \le q} \beta_{ij} x_i x_j. \tag{1.12}$$

Any mixture experiment with expected response, when studied using K-models is homogeneous in ingredients. The mixture ingredients, x_i can conveniently be written as a $q \times 1$ vector $\boldsymbol{x} = (x_1, x_2, \ldots, x_q)'$.

The Kronecker product $x \otimes x$ consists of q^2 cross products arranged lexicographically as follows:

$$x \otimes x = (x_1x_1, x_1x_2, ..., x_1x_q, \; x_2x_1, ..., x_2x_q, ..., x_qx_1, ..., x_qx_q)' \tag{1.13}$$

The Kronecker product $x \otimes x \otimes x$ consists of q^3 cross products arranged lexicographically such that

$$x \otimes x \otimes x = (x_1x_1x_1, x_1x_1x_2, ..., x_1x_1x_q, x_1x_2x_2, ..., x_1x_2x_q, ..., x_qx_qx_1, ..., x_qx_qx_q)' \tag{1.14}$$

The K-models are given as

$$\eta = \sum_{1 \le i, j \le q} \theta_{ij} x_i x_j$$

$$\eta = \sum_{1 \le i, j, k \le q} \theta_{ijk} x_i x_j x_k$$

Draper and Pukelshiem (1999) studied the Kiefer design ordering of simplex designs for first and second degree mixture models by discussing the improvement of a given design in terms of increasing symmetry as well as obtaining a larger moment matrix under the Loewner ordering of matrices. The two criteria together define the Kiefer design ordering. Draper and Pukelshiem (1999) showed that for the second-degree mixture model, the set of weighted centroid designs constitutes a convex complete class for the Kiefer ordering. For four ingredients, the class is minimal complete and for $q \geq 5$, the set of weighted centroid design is complete. Klein (2004) studied optimal weighted centroid designs for second degree Kronecker model mixture experiments.

An excellent survey article on known results about analytic solutions and numerical solutions of optimal designs for various regression models for experiments with mixtures has been carried out by Chan (2000). Cornell (2002) presented an exhaustive coverage of designs and alternative model forms. Piepel and Cornell (2008) have presented an excellent reference of application papers using mixture designs. Mixture experiments have been applied in the following fields, viz., food, rubber, plastics, ceramics, industrial chemistry, cereal chemistry, chemical ecology, detergent, glass, paint, pharmaceutics industry, physical and engineering sciences, nuclear, liquid chromatography and hazardous waste management, heat and material balance problems, among others.

1.7. Process Variables in Mixture Experiments

Process variables are those factors present in an experiment that do not physically form any part of the mixture but whose levels, when altered, may affect the blending of the ingredients. In the mixture experiment consisting of q components, we may also have n process variables. For example, in making sandwich patties, the time of deep fat frying, cooking oven temperature and cooking time could serve as the three process variables (Cornell (2002)).

The experimental design for a mixture problem involving process variables was considered as a factorial by Gorman and Cornell (1982), while Daniel (1963) considered it as a fractional factorial design in the process variables at each point in a lattice in the mixture simplex. Cornell and Gorman (1984) presented combined mixture component–process variable (MPV) designs for $n \geq 3$ that use only a fraction of the total number of design points. Cornell (1988) and Hare (1979) suggested the split plot designs and Vuchkov et al. (1981) considered a quadratic model that contained interaction terms between the mixture components and process variables. Wynn (1970) used Wynn's algorithm to generate 10 discrete quasi D-optimal designs for 3 to 6 mixture components and 1 to 4 process variables. Nigam (1970, 1976) and Saxena and Nigam (1973) used orthogonal blocking to divide the mixture blends into subsets called blocks such that when the required model is fitted, the linear and quadratic terms in the model are orthogonal to the blocks. John (1984) considered block designs in which design points at the same value of the process variables are grouped into the same block. John (1984) used Box and Hunter's (1957) orthogonality conditions and obtained orthogonal block designs based on Latin squares for mixture experiments involving process variables.

Næs, Færgestad and Cornell (1998) compared methods for analyzing data from a three component mixture experiment in the presence of variation created by two process variables. Cornell (1995, 2002), Piepel and Cornell (1985, 1987) and Myers and Montgomery (2002) discussed mixture-process variable experiments. Kowalski et al. (2000) proposed a new model for mixture experiments involving process variables. Chantarat et al. (2006) presented a combined array approach to minimize expected prediction errors in experiments involving mixture and process variables. An optimal design for mixture-process experiments involving control and noise variables was studied by Chung et al. (2007).

1.8. Orthogonal Block Designs for Mixture Experiments

Groups of mixture blends where each group or block is assumed to differ from the other groups or blocks by an additive constant are called block designs. If the estimates of the blending properties in the fitted model are uncorrelated with and are unaffected by the effects of the blocks then the design is said to block orthogonally with respect to the blending properties of the components. In mixture experiments, we may run one or more of the blocks at each combination of the process variables.

Let N mixture blends (not necessarily all distinct) be arranged in t blocks such that the w^{th} block contains n_w blends and $n_1 + n_2 + \cdots + n_t = N$. In this case the model is

$$E(y) = \sum_{i=1}^{q}\beta_i x_i + \sum_{1\le i<j\le q}\beta_{ij}x_i x_j + \sum_{w=1}^{t-1}\gamma_w Z_w. \tag{1.15}$$

where Z_w is a dummy block variable and γ_w is the block difference parameter for w = 1, 2, … , t-1 with t as the number of blocks.

Nigam (1970, 1976) defined conditions for the estimation of the parameters of Scheffé’s quadratic model in the presence of block effects and constructed designs which satisfy these blocking conditions. John (1984) gave the following conditions for the orthogonal block of mixture blends

$$\sum_{u=1}^{n_w} x_{ui} = k_{1w} \qquad \text{(constant);} \qquad i = 1, 2, \ldots, q$$

$$\sum_{u=1}^{n_w} x_{ui} x_{uj} = k_{2w} \qquad \text{(constant);} \qquad 1 \leq i < j \leq q \tag{1.16}$$

In particular, it is necessary that in each block the total of the volume fractions for the first component shall be k_1 and the total for the second component shall be k_2; it is not necessary that $k_1 = k_2$.

We consider designs in which the least squares estimators of the mixture parameters are uncorrelated with those of the block parameters, i.e., the blocks are orthogonal. The concept of orthogonal blocks can be conveniently defined in terms of the extended design matrix $\boldsymbol{X}$ for mixture parameters and the design matrix $\boldsymbol{Z}$ for the blocking variables so that the full model is

$$E(\text{y}) = \boldsymbol{X}\beta + \boldsymbol{Z}\gamma \tag{1.17}$$

where β and γ are the column vectors of constant coefficients. The design is orthogonally blocked if and only if $\boldsymbol{X}'\boldsymbol{Z} = \mathbf{0}$ or $\boldsymbol{Z}'\boldsymbol{X} = \mathbf{0}$.

John (1984) defined blocking arrangements based on orthogonal latin squares starting with standard latin squares for four component mixture designs. A latin square with both its first row and first column in the order *a*, *b*, *c*, *d* is said to be in standard order. Table 1.3 lists the four standards latin squares of order 4.

Table 1.3. Standard Latin squares of Order 4

Square 1	Square 2	Square 3	Square 4
a b c d	*a b c d*	*a b c d*	*a b c d*
b a d c	*b d a c*	*b c d a*	*b a d c*
c d a b	*c a d b*	*c d a b*	*c d b a*
d c b a	*d c b a*	*d a b c*	*d c a b*

Each of these standard latin squares yield six squares (including the original square) by permuting the last three columns. For example, if we consider Square 4 and permute the second, third and fourth columns, we obtain the squares as shown in Table 1.4.

Table 1.4. Six Squares obtained by permuting the last three columns

S I	S II	S III
a b c d	*a c d b*	*a d b c*
b a d c	*b d c a*	*b c a d*
c d b a	*c b a d*	*c a d b*
d c a b	*d a b c*	*d b c a*

S IV	S V	S VI
a b d c	*a d c b*	*a c b d*
b a c d	*b c d a*	*b d a c*
c d a b	*c a b d*	*c b d a*
d c b a	*d b a c*	*d a c b*

For all the six squares, $\sum_{u=1}^{4} x_{ui}x_{uj} = \sum x_{uk}x_{ul} = 2(ab+cd)$, and the remaining four cross products add to a different constant, that is, $\sum x_{um}x_{up} = ac+bd+bc+ad$, $i < j$, $k < l$, $m < p$. Take square S I for example, if $i = 1$, then $j = 2$ and if $k = 3$ then $l = 4$. Also, if $m = 1$ or 2, then $p = 3$ or 4. The same is true for square S IV. Squares S I and S IV are called “mates” since for both these squares, $k_{12} = k_{34}$ and $k_{13} = k_{14} = k_{23} = k_{24}$.

Table 1.5. Portion of *X* Matrix along with the column sums arising from S I

	1	2	3	4	12	13	14	23	24	34
	a	*b*	*c*	*d*	*ab*	*ac*	*ad*	*bc*	*Bd*	*cd*
	b	*a*	*d*	*c*	*ab*	*bd*	*bc*	*ad*	*Ac*	*cd*
	c	*d*	*b*	*a*	*cd*	*bc*	*ac*	*bd*	*Ad*	*ab*
	d	*c*	*a*	*b*	*cd*	*ad*	*bd*	*ac*	*Bc*	*ab*
Column Sum	1	1	1	1	A	B	B	B	B	A

Table 1.5 and Table 1.6 present the portion of ***X*** Matrix along with the column sums arising from S I and S IV, respectively.

Table 1.6. Portion of *X* Matrix along with the column sums arising from S IV

	1	2	3	4	12	13	14	23	24	34
	a	*b*	*d*	*c*	*ab*	*ad*	*ac*	*bd*	*bc*	*cd*
	b	*a*	*c*	*d*	*ab*	*bc*	*bd*	*ac*	*ad*	*cd*
	c	*d*	*a*	*b*	*cd*	*ac*	*bc*	*ad*	*bd*	*ab*
	d	*c*	*b*	*a*	*cd*	*bd*	*ad*	*bc*	*ac*	*ab*
Column Sum	1	1	1	1	A	B	B	B	B	A

where

$a + b + c + d = 1$,

A = 2($ab + cd$),

B = $ac + bd + ad + bc$

By adding one common additional blend (the centroid) to each of the two blocks, we obtain orthogonal block design based on latin squares.

	BLOCK I					BLOCK II			
S I	*a*	*b*	*c*	*d*	S IV	*a*	*b*	*d*	*c*
	b	*a*	*d*	*c*		*b*	*a*	*c*	*d*
	c	*d*	*b*	*a*		*c*	*d*	*a*	*b*
	d	*c*	*a*	*b*		*d*	*c*	*b*	*a*
S III	*a*	*d*	*b*	*c*	S VI	*a*	*c*	*b*	*d*
	b	*c*	*a*	*d*		*b*	*d*	*a*	*c*
	c	*a*	*d*	*b*		*c*	*b*	*d*	*a*
	d	*b*	*c*	*a*		*d*	*a*	*c*	*b*
	1/4	1/4	1/4	1/4		1/4	1/4	1/4	1/4

Mates are pairs of squares that satisfy the orthogonality conditions given in (1.16). The pairs of mates are given below:

Square 2: Mates are (S I and S VI), (S II and S IV), (S III and S V).

Square 3: Mates are (S I and S V), (S II and S VI), (S III and S IV).

Square 4: Mates are (S I and S IV), (S II and S V), (S III and S VI).

No mates exist for Square 1.

Czitrom (1988, 1989, 1992) used John's (1984) designs and obtained D-optimal orthogonal designs in three and four components. Draper *et al*. (1993) studied mixture designs for four components in orthogonal blocks, which were extended by Prescott *et* *al*. (1993) for five components. Lewis *et al*. (1994) presented general methods of constructing designs for q mixture components in two or more orthogonal blocks using latin squares. Prescott *et al*. (1997) obtained D-optimal mixture designs for five components in two orthogonal blocks. Chan and Sandhu (1999) discussed A- and E-optimal orthogonal block designs for three component mixture experiments. A-optimal orthogonal block designs for four component mixture experiments were obtained by Ghosh and Liu (1999).

D-, A- and E-optimal orthogonal designs in two blocks with three and four components for Becker's mixture models were obtained by Aggarwal *et al*. (2002). Singh (2003) obtained D-, A- and E-optimal orthogonal designs in two blocks for Darroch and Waller's quadratic mixture model in three and four components. Aggarwal *et al*. (2004) presented D-, A- and E-optimal orthogonal block designs in two blocks for second degree K-model in three or four components. D-optimal experimental designs in two orthogonal blocks for Darroch and Waller's quadratic model in constrained mixture components were obtained by Aggarwal and Singh (2006).

1.9. F-Squares

F-squares have been studied by Finney (1945, 1946a, 1946b), Freeman (1966) and Addleman (1967). Hedayat et al. (1975) made further contributions to the theory of F-squares. Laywine (1989) obtained F-squares by making substitutions based on numbers for latin squares. F-squares and orthogonal F-squares are a generalization of latin squares and orthogonal latin squares, respectively. Hedayat and Seiden (1970) gave the following definition:

***Definition* 1.1:** Let $\mathbf{A} = [a_{ij}]$ be an $n \times n$ matrix and let $\boldsymbol{\Sigma} = (c_1, c_2, \ldots, c_m)$ be the ordered set of distinct elements of $\mathbf{A}$. In addition, suppose that for each $k = 1, 2, ..., m$, c_k appears precisely λ_k times ($\lambda_k \geq 1$) in each row and in each column of $\mathbf{A}$. Then, $\mathbf{A}$ will be called a frequency square or more concisely, an F-square of order n on $\boldsymbol{\Sigma}$ with frequency vector $(\lambda_1, \lambda_2, \ldots, \lambda_m)$ and is denoted by $\mathrm{F}(n; \lambda_1, \lambda_2, \ldots, \lambda_m)$.

Laywine (1989) studied F-squares by making substitutions on the symbols of latin squares. For example, consider the following latin square of order 4. By substituting the symbol $d = a$ in the latin square, we obtain F (4; 2, 1, 1) defined on $\boldsymbol{\Sigma} = (a, b, c)$. Aggrawal et al. (2009) denoted this F-square as FSI(4), where 4 in the parenthesis denotes the number of components.

Latin Square of order 4				**FSI(4) Square number 1**				**FSI(4) Square number 2**				**FSI(4) Square number 3**			
a	*b*	*c*	*d*	*a*	*b*	*c*	*a*	*a*	*c*	*a*	*b*	*a*	*a*	*b*	*c*
b	*a*	*d*	*c*	*b*	*a*	*a*	*c*	*b*	*a*	*c*	*a*	*b*	*c*	*a*	*a*
c	*d*	*a*	*b*	*c*	*a*	*a*	*b*	*c*	*a*	*b*	*a*	*c*	*b*	*a*	*a*
d	*c*	*b*	*a*	*a*	*c*	*b*	*a*	*a*	*b*	*a*	*c*	*a*	*a*	*c*	*b*

FSI(4) generates two distinct F-squares, namely Square number 2 and Square number 3 via permutations of the last three columns. Aggrawal et al. (2009) identified F-squares by simply writing down the first row of the square and represented by Square number 2 by writing its first row as *a c a b*. Aggrawal et al. (2009) gave the following definitions.

***Definition* 1.2:** An F-square with the first row and first column in natural order is called a standard F-square where by natural order we mean to imply that each element is followed by the same element (if it assumes an equal proportion) or the next element cyclically. For example, for four components if $d = a$, then $\boldsymbol{\Sigma} = (a, b, c)$, the order could either be a, a, b, c or a, b, c, a.

***Definition* 1.3:** Two F-squares are equivalent if one can be derived from the other by permutations of rows and/or permutations of columns and/or permutations of elements.

***Definition* 1.4:** Two F-squares are conjugates if the rows of one are the columns of the other.

***Definition* 1.5:** Distinct F-squares of order q are mates if they have identical cross product sums, i.e., if the columns of the two distinct F-squares have identical sums of the inner product of columns.

Aggarwal et al. (2008) obtained optimal orthogonal block designs in two blocks based on F-squares for Darroch and Waller's quadratic mixture model in four components. Aggarwal et al. (2009) constructed mixture designs in orthogonal block designs based on F-squares and presented a general algorithm to obtain the mates of F-squares.

Aggarwal et al. (2011a) presented nearly optimal orthogonally blocked designs for four mixture components based on F-square designs. Aggarwal et al. (2011b) presented orthogonally blocked mixture component-amount designs via projections of F-squares. Aggarwal et al. (2013) obtained optimal orthogonal block designs for four mixture components in two blocks for Becker's models and K-model.

1.10. Uniform Designs and Uniformity Measures

If the model is known to be of a particular form, such as a quadratic or a cubic, then optimality considerations suggest that some of the design points should be pushed as far out as possible to the boundaries of the available design space. McKay et al. (1979) proposed a method of generating a set of experimental points $\mathcal{P} = \{x_1, x_2, \ldots, x_n\}$ called latin hypercube sampling. The uniform design (UD) proposed by Fang (1980) is the application of number-theoretic methods in experimental designs with uniform distributions. Fang and Wang (1981) employed the good lattice point method in Quasi-Monte Carlo methods. Fang et al. (1999) proposed a construction method based on latin squares. Fang et al. (2000) suggested a way for constructing nearly uniform designs and orthogonal designs. Fang and Winker (1998) obtained U-type optimal design in Monte Carlo and Quasi-Monte Carlo methods by Niederreiter and Peart (1986). Fang et al. (2001) employed a powerful construction of uniform design viz., the latin hypercube design and the Centered L_2-discrepancy of random sampling.

A set of the form $\mathcal{P}_n = \{x_k = (x_{k1}, \ldots, x_{ks})\ ,\ k = 1, \ldots, n\}$ is called a *lattice point set*, where $\{x\}$ denotes the fractional part of x and n is an integer ($n \geq 2$) and $\mathbf{a} = (a_1, \ldots, a_s)$ an integer vector modulo n. The vector $\mathbf{a}$ is called a lattice point set. The *glp* set is obtained by a so-called good lattice point modulo n.

***Definition* 1.6:**

Let $(n; h_1, \ldots, h_s)$ be a vector of integers satisfying $1 \leq h_i < n$, $h_i \neq h_j$ for $i \neq j$, $s < n$ and the greatest common divisors $(n, h_i) = 1$, $i = 1, \ldots, s$. Let

$$q_{ki} = kh_i \ (\mathrm{mod}\ n)$$

$$x_{ki} = (2q_{ki} - 1) / 2n, \quad k = 1, \ldots, n, \quad i = 1, \ldots, s. \tag{1.18}$$

If the set $\mathcal{P}_n$ has the smallest discrepancy among all possible generating vectors, then the set $\mathcal{P}_n$ is called a *glp* set. It can be seen that x_{ki} defined in (1.18) can be easily calculated by

$$x_{ki} = \left\{ \frac{2kh_i - 1}{2n} \right\} \tag{1.19}$$

where $\{ x \}$ stands for the fractional part of x.

***Example* 1.1:** Take $n = 7$, $s = 3$, $h_1 = 1$, $h_2 = 3$ and $h_3 = 6$. We have

$$(q_{ki}) = \begin{pmatrix} 1 & 3 & 6 \\ 2 & 6 & 5 \\ 3 & 2 & 4 \\ 4 & 5 & 3 \\ 5 & 1 & 2 \\ 6 & 4 & 1 \\ 7 & 7 & 7 \end{pmatrix} \qquad \begin{aligned} x_1 &= (1/14, 5/14, 11/14) \\ x_2 &= (3/14, 11/14, 9/14) \\ x_3 &= (5/14, 3/14, 7/14) \\ x_4 &= (7/14, 9/14, 5/14) \\ x_5 &= (9/14, 1/14, 3/14) \\ x_6 &= (11/14, 7/14, 1/14) \\ x_7 &= (13/14, 13/14, 13/14) \end{aligned}$$

and $\{x_k, k = 1, \ldots, 7\}$ is the lattice point set of the generating vector (7; 1, 3, 6).

Niederreiter (1978, 1988 and 1992) recommended Quasi-Monte Carlo methods for multi-dimensional numerical integration with pseudo-random numbers. Shaw (1988) used the glp set for comparing among several methods defined by Niederreiter (1978, 1988).

The Koksma-Hlawka inequality gives the upper error bounds of the estimate of $E\{h(x)\}$ (Hua and Wang (1981)).

$$|E\{h(x)\} - h| \leq D(\mathcal{P})V(h) \tag{1.20}$$

where $V(h)$ is a measure of variation of h and $D(\mathcal{P})$ is the discrepancy of $\mathcal{P}$, a measure of the uniformity of $\mathcal{P}$.

Let $\mathcal{P} = \{x_1, \ldots, x_n\}$ be set of n points in the s-dimensional unit cube $C^s = [0, 1]^s$. Many different measures of uniformity of $\mathcal{P}$ have been defined by Fang and Wang (1994), Hickernell (1998a, 1998b) and Hickernell et al. (2000)).

Let F(x) be the uniform distribution on C^s and $F_n(\boldsymbol{x})$ be the empirical distribution function of $\boldsymbol{X}$, i.e.,

$$F_n(x) = \frac{1}{n}\sum_{i=1}^{n} I\{x_i \le x\} \tag{1.21}$$

where I{.} is the indicator function and all inequalities are understood to be component wise. The L_p-discrepancy of $\mathcal{P}$ is defined as

$$D_p(\mathcal{P}) = \left[\int_{C^s} |F_n(x) - F(x)|^p dx\right]^{\frac{1}{p}} \tag{1.22}$$

where $F(\boldsymbol{x})$ is the distribution function of the uniform distribution over C^s. When $p = \infty$, $D \equiv D_\infty$ is also called the discrepancy (or star-discrepancy). This is probably the most commonly used measure of discrepancy and can further be expressed as

$$D(\mathcal{P}) = sup_{x\in C^s} |F_n(x) - F(x)| \tag{1.23}$$

This discrepancy has been universally accepted in Quasi-Monte Carlo methods and number-theoretic methods. Bundschuh and Zhu (1993) presented a method for exact calculation of the discrepancy of low dimensional finite point sets. Warnock (1972) gave the following analytical expression for calculating L_2-discrepancy

$$\left(D_2(\mathcal{P})\right)^2 = 3^{-s} - \frac{2^{1-s}}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}(1 - x_{kl}^2) + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left[1 - max\left(x_{ki}, x_{ji}\right)\right] \tag{1.24}$$

where $x_k = (x_{k1}, \ldots, x_{ks})$, $\mathcal{P} = \{x_1, x_2, \ldots, x_n\}$ be a set of n points in the s-dimensional unit cube $C^s = [0, 1]^s$.

Hickernell (1998a) gave three modified L_2-discrepancies: the symmetric L_2-discrepancy (SD_2), the centered L_2-discrepancy (CD_2) and modified L_2-discrepancy (MD_2). Ma (1997a, 1997b) obtained a number of good properties of the symmetrical discrepancy. These uniformity measures are described in Fang and Mukherjee (2000).

Hickernell (1998a) gave an analytical expression for the centered L_2-discrepancy is

$$\begin{aligned}\left(CD_2(\mathcal{P})\right)^2 = \left(\frac{13}{12}\right)^s &- \frac{2}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}\left(1 + \frac{1}{2}|x_{kl} - 0.5| - \frac{1}{2}|x_{kl} - 0.5|^2\right) \\ &+ \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(1 + \frac{1}{2}|x_{ki} - 0.5| + \frac{1}{2}\left|x_{ji} - 0.5\right| - \frac{1}{2}\left|x_{ki} - x_{ji}\right|\right)\end{aligned} \tag{1.25}$$

The wrap-around L_2-discrepancy proposed by Hickernell (1998b) has the following analytical form.

$$\left(WD_2(\mathcal{P})\right)^2 = -\left(\frac{4}{3}\right)^s + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(\frac{3}{2} - \left|x_{ki} - x_{ji}\right|\left(1 - \left|x_{ki} - x_{ji}\right|\right)\right) \quad (1.26)$$

where $x_k = (x_{k1}, \dots, x_{ks}) \in \mathcal{P}$, the centered L_p-discrepancy takes into account not only the uniformity of $\mathcal{P}$ over C^s, but also the uniformity of all the projections of $\mathcal{P}$ over C^s.

Fang and Wang (1981) presented a note on uniform distribution and experimental design. They gave the following formula to compute the discrepancy of the G-Uniform design.

$$D(n,a) = \frac{1}{n}\sum_{k=1}^{n}\prod_{v=1}^{s}\left(1 - \frac{2}{\pi} ln\left(2 sin\pi \frac{a_{vk}}{n+1}\right)\right) \quad (1.27)$$

where $a_{vk} = a_v k (\text{mod}\, n)$, $1 \leq a_{vk} < n$ for $1 \leq k < n$ and $a_{vn} = n$.

Uniform design allocates experimental points that are uniformly scattered on the domain in the sense of low-discrepancy (Fang and Wang (1994)).

1.11. Conclusions

This thesis contains five chapters. The objective is to obtain optimal orthogonally blocked mixture designs for inverse model, reduced cubic canonical model, F-square based uniform designs and efficient uniform designs in three and four mixture components. A comprehensive bibliography has also been given at the end, which has been referred to during the research work.

In the ***first Chapter,*** basic ideas about the general mixture problems for understanding the different concepts as regards mixture experiments and uniform designs with low discrepancy are discussed.

In ***Chapter 2,*** we have obtained F-square based D-, A-, and E-optimal orthogonally blocked designs in four mixture components for inverse models

In ***Chapter 3,*** focuses on D-, A- and E-optimal designs and optimal orthogonally blocked designs based on F-squares for reduced cubic canonical model in four mixture components.

In ***Chapter 4,*** we have obtained uniform designs based on cyclic F-squares with low discrepancy.

In ***Chapter 5,*** we have discussed centered L_2-discrepancy and have obtained D-, A- and G-efficient uniform designs for mixture experiments in three and four components based on F-squares designs.

Chapter 2

FOUR COMPONENT OPTIMAL ORTHOGONALLY BLOCKED DESIGNS BASED ON F-SQUARES FOR MIXTURE INVERSE MODEL

2.1. Introduction

In mixture experiments, the response is assumed to be a function of the proportions of the q components present in the mixture and is independent of the total amount of the mixture. The factor space is depicted by the following ($q - 1$) dimensional simplex:

$$S_{q-1} = \left\{ x = \left(x_1, x_2, ..., x_q\right) \mid \sum_{i=1}^{q} x_i = 1,\ x_i \geq 0 \right\} \tag{2.1}$$

where x_i (i = 1, 2, …, q) denotes the components of the mixture. Some factors known as process variables are present in experiments. The process variables do not constitute any portion of the mixture but their different levels may significantly affect the blending properties of the ingredients. For example, the driving speed and automobile size may affect the blending behaviors of fuels being tested to compare the average mileage of the fuels individually as well as when blended together, (Cornell (2002)).

Draper and St. John (1977) presented the following mixture model with inverse terms for modeling extreme changes in the response as the value of certain components tends to the boundary (usually the ‘ zero’ boundary).

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{-i} x_i^{-1}. \tag{2.2}$$

The model is an augmentation of the Scheffé’s polynomials with the additional terms of the form x_i^{-1} included to account for the possible edge effects (Draper and St. John (1977)) in the response as x_i approaches zero. It is assumed that the value of x_i never reaches zero, but that the value could be very close to zero; i.e., $x_i \rightarrow \varepsilon_i > 0$; where ε_i is a very small positive quantity. The following equation presents the Taylor’s series expansion of x_i^{-1}

$$1/x_i = 1 - (x_i - 1) + (x_i - 1)^2 - (x_i - 1)^3 + (x_i - 1)^4 - \cdots \tag{2.3}$$

contains terms in $x_i,\ x_i^2,\ x_i^3,\ \ldots$. Here x_i^{-1} may be regarded as a special form of polynomial.

John (1984) gave simple conditions for orthogonal blocking of blends for Scheffé’s quadratic model and constructed latin square based designs satisfying those conditions. Aggarwal et al. (2009) constructed orthogonal block designs based on F-squares for Scheffé's quadratic model and obtained D-, A- and E-optimal designs for $q = 4$. Aggarwal et al. (2008) obtained D-, A- and E-optimal orthogonal block designs in two blocks based on F-squares for Darroch and Waller's quadratic mixture model in four components. In particular for $q = 4$, Aggarwal et al. (2009) obtained the D-, A- and E-optimalities of F-square based designs.

In this chapter, we give conditions for the orthogonal blocking of blends for Draper and St. John's (1977) quadratic model with inverse terms and construct D-, A- and E-optimal orthogonal block designs in four components for the classes of designs that satisfy the blocking conditions for Draper and St. John's (1977) quadratic model having inverse terms.

2.2. Blocking Conditions

When m mixture blends (not necessarily all distinct) are arranged in two blocks B_1 and B_2 with m_1 and m_2 blends respectively and $m_1 + m_2 = m$, the model (2.2) with block effect is γ

$$E(y) = \sum_{i=1}^{q} \beta_i x_{iu} + \sum_{i=1}^{q} \beta_{-i} x_{iu}^{-1} + \gamma Z_u + e_u \;;\; u = 1, 2, ..., m \tag{2.4}$$

Here $Z_u = -1$, for the blends in block B_1 and $Z_u = 1$, for the blends in block B_2 and e_u's are random errors which are independently distributed with mean 0 and same variance σ^2. Model (2.4) does not contain the product terms of x_i and Z_u. We may express model given in (2.4) in matrix notation as

$$E(y) = \boldsymbol{X\beta} + \boldsymbol{Z\gamma} \tag{2.5}$$

where $\boldsymbol{X}$ is the $m \times 2q$ matrix corresponding to the mixture part, $\boldsymbol{\beta}$ is the $2q \times 1$ column vector of unknown parameters, $\boldsymbol{\gamma}$ is the block effect parameter and $\boldsymbol{Z}$ is the $m \times 1$ column vector corresponding to the block variable $\boldsymbol{Z}$. To ensure that the two blocks of mixture blends will be orthogonal in the sense that the block effects do not affect the estimates of the coefficients in the mixture inverse model, we satisfy the condition that $\boldsymbol{X'Z} = \boldsymbol{0}$. We have obtained the following conditions for the orthogonal blocking for Draper and St. John's (1977) Inverse model.

$$\sum_{u=1}^{m_w} x_{ui} = k_i,\ \sum_{u=1}^{m_w} x_{ui}^{-1} = k_i' \ \ \forall\, w = 1,2 \ \text{ and } \ i = 1,2,\dots,q \tag{2.6}$$

where k_i's and k_i''s are constants (i = 1, 2, …, q).

We construct F-squares based orthogonally blocked four component mixture designs involving inverse terms and achieve orthogonal blocking of blends by satisfying the conditions given in (2.6).

2.3. Orthogonally Blocked Four Component Mixture Designs with Inverse Terms using F-Squares

For four component mixtures, nine distinct runs are required to estimate all the parameters in the inverse model (2.4). With a single block variable at two levels, $Z = -1$ and $Z = +1$, we may take one block at $Z = -1$ and the other block at $Z = +1$. Aggarwal et al. (2009) suggested the class of designs given below that are based on F-squares with an added observation at the centroid and a, b, c are numbers between 0 to 1 such that $2a + b + c = 1$. There are 18 runs in two blocks in each design. Each block contains 9 runs representing the specific four component mixtures.

Aggarwal et al. (2009) suggested the class of designs given in (2.7), (2.8) and (2.9), respectively. This class of designs consists of 13 distinct quaternary blends. We use this class of designs to obtain the D-, A- and E-optimal orthogonal block designs for Draper and St. John's (1977) mixture model with inverse terms in four components.

DESIGN 1:

$$B_1 = \left|\begin{matrix} a & b & c & a \\ b & c & a & a \\ c & a & a & b \\ a & a & b & c \\ a & c & a & b \\ b & a & a & c \\ c & a & b & a \\ a & b & c & a \\ 1/4 & 1/4 & 1/4 & 1/4 \end{matrix}\right| \text{ and } B_2 = \left|\begin{matrix} a & a & c & b \\ b & a & a & c \\ c & b & a & a \\ a & c & b & a \\ a & c & b & a \\ b & a & c & a \\ c & a & a & b \\ a & b & a & c \\ 1/4 & 1/4 & 1/4 & 1/4 \end{matrix}\right| \tag{2.7}$$

DESIGN 2:

$$B_1 = \left|\begin{array}{cccc} a & b & c & a \\ b & c & a & a \\ c & a & a & b \\ a & a & b & c \\ a & a & b & c \\ b & a & c & a \\ c & b & a & a \\ a & c & a & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{array}\right| \text{ and } B_2 = \left|\begin{array}{cccc} a & a & c & b \\ b & a & a & c \\ c & b & a & a \\ a & c & b & a \\ a & b & a & c \\ b & c & a & a \\ c & a & b & a \\ a & a & c & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{array}\right| \tag{2.8}$$

DESIGN 3:

$$B_1 = \left|\begin{array}{cccc} a & c & a & b \\ b & a & a & c \\ c & a & b & a \\ a & b & c & a \\ a & a & b & c \\ b & a & c & a \\ c & b & a & a \\ a & c & a & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{array}\right| \text{ and } B_2 = \left|\begin{array}{cccc} a & c & b & a \\ b & a & c & a \\ c & a & a & b \\ a & b & a & c \\ a & b & a & c \\ b & c & a & a \\ c & a & b & a \\ a & a & c & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{array}\right| \tag{2.9}$$

The form of matrix $\boldsymbol{X}$ for Design 1 is as follows for model (2.2).

$$\boldsymbol{X}=\begin{bmatrix}
a & b & c & a & \frac{1}{a} & \frac{1}{b} & \frac{1}{c} & \frac{1}{a} \\
b & c & a & a & \frac{1}{b} & \frac{1}{c} & \frac{1}{a} & \frac{1}{a} \\
c & a & a & b & \frac{1}{c} & \frac{1}{a} & \frac{1}{a} & \frac{1}{b} \\
a & a & b & c & \frac{1}{a} & \frac{1}{a} & \frac{1}{b} & \frac{1}{c} \\
a & c & a & b & \frac{1}{a} & \frac{1}{c} & \frac{1}{a} & \frac{1}{b} \\
b & a & a & c & \frac{1}{b} & \frac{1}{a} & \frac{1}{a} & \frac{1}{c} \\
c & a & b & a & \frac{1}{c} & \frac{1}{a} & \frac{1}{b} & \frac{1}{a} \\
a & b & c & a & \frac{1}{a} & \frac{1}{b} & \frac{1}{c} & \frac{1}{a} \\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 4 & 4 & 4 & 4 \\
a & a & c & b & \frac{1}{a} & \frac{1}{a} & \frac{1}{c} & \frac{1}{b} \\
b & a & a & c & \frac{1}{b} & \frac{1}{a} & \frac{1}{a} & \frac{1}{c} \\
c & b & a & a & \frac{1}{c} & \frac{1}{b} & \frac{1}{a} & \frac{1}{a} \\
a & c & b & a & \frac{1}{a} & \frac{1}{c} & \frac{1}{b} & \frac{1}{a} \\
a & c & b & a & \frac{1}{a} & \frac{1}{c} & \frac{1}{b} & \frac{1}{a} \\
b & a & c & a & \frac{1}{b} & \frac{1}{a} & \frac{1}{c} & \frac{1}{a} \\
c & a & a & b & \frac{1}{c} & \frac{1}{a} & \frac{1}{a} & \frac{1}{b} \\
a & b & a & c & \frac{1}{a} & \frac{1}{b} & \frac{1}{a} & \frac{1}{c} \\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 4 & 4 & 4 & 4
\end{bmatrix}$$

The forms of $\boldsymbol{X'X}$ for Design 1, Design 2 and Design 3 are as given in (2.10), (2.11) and (2.12), respectively. The two blocks in all the three designs satisfy the orthogonality conditions given in (2.6). Moreover, since the blocks in Design 1, Design 2 and Design 3 are orthogonal, it is unnecessary to consider the block variable *Z* while optimizing the mixture design and we need to focus on matrix ***X′X*** only.

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccc} P & R & R & Q & S & U & U & T \\ R & P & Q & R & U & S & T & U \\ R & Q & P & R & U & T & S & U \\ Q & R & R & P & T & U & U & S \\ \hline S & U & U & T & X & Z & Z & Y \\ U & S & T & U & Z & X & Y & Z \\ U & T & S & U & Z & Y & X & Z \\ T & U & U & S & Y & Z & Z & X \end{array}\right| \qquad (2.10)$$

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccc} P & Q & R & R & S & T & U & U \\ Q & P & R & R & T & S & U & U \\ R & R & P & Q & U & U & S & T \\ R & R & Q & P & U & U & T & S \\ \hline S & T & U & U & X & Y & Z & Z \\ T & S & U & U & Y & X & Z & Z \\ U & U & S & T & Z & Z & X & Y \\ U & U & T & S & Z & Z & Y & X \end{array}\right| \qquad (2.11)$$

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccc} P & R & Q & R & S & U & T & U \\ R & P & R & Q & U & S & U & T \\ Q & R & P & R & T & U & S & U \\ R & Q & R & P & U & T & U & S \\ \hline S & U & T & U & X & Z & Y & Z \\ U & S & U & T & Z & X & Z & Y \\ T & U & S & U & Y & Z & X & Z \\ U & T & U & S & Z & Y & Z & X \end{array}\right| \qquad (2.12)$$

where,

$P = 1/8 + 8a^2 + 4b^2 + 4c^2$ $\qquad$ $Q = 1/8 + 8a^2 + 4ab + 4ac + 4bc$

$R = 1/8 + 2a^2 + 6ab + 6ac + 2bc$ $\qquad$ $S = 18$

$$T = 6+\frac{2a}{b}+\frac{2b}{a}+\frac{2a}{c}+\frac{2b}{c}+\frac{2c}{a}+\frac{2c}{b} \qquad U = 4+\frac{3a}{b}+\frac{3b}{a}+\frac{3a}{c}+\frac{b}{c}+\frac{3c}{a}+\frac{c}{b}$$

$$X = 32+\frac{8}{a^2}+\frac{4}{b^2}+\frac{4}{c^2} \qquad Y = 32+\frac{4}{a^2}+\frac{4}{ab}+\frac{4}{ac}+\frac{4}{bc}$$

$$Z = 32+\frac{2}{a^2}+\frac{6}{ab}+\frac{6}{ac}+\frac{2}{bc}$$

2.4. Optimal Designs for Inverse Model

In order to obtain D-, A- and E-optimal designs for the Inverse Model given in (2.4), we obtain the expressions for $|\boldsymbol{X'X}|$, T = Trace $(\boldsymbol{X'X})^{-1}$ and the eigenvalues λ_i (i = 1, 2, …, 8) of $\boldsymbol{X'X}$ which are as given in (2.13), (2.14) and (2.15), respectively.

$$|\boldsymbol{X'X}| = \frac{4608(a-b)^6\,(b-c)^6\,(a-c)^6\,(-ab+16abc^2-ac-2bc+32a^2bc+16ab^2c)^2}{a^8b^8c^8} \tag{2.13}$$

$$T = T_1 / T_2 \tag{2.14}$$

$$\begin{aligned}
T_1 = (&11a^4b^4+24a^6b^4-48a^5b^5+24a^4b^6+14a^4bc^3+30a^3bc^4-656a^5bc^4-448a^4bc^5+48a^3bc^6\\
&+14a^4b^3c+30a^3b^4c-656a^5b^4c-448a^4b^5c+48a^3b^6c+6a^4b^2c^2-48a^6b^2c^2-30a^3b^3c^2\\
&-192a^5b^3c^2+2a^2b^4c^2\ 2-888a^4b^4c^2+12049a^6b^4c^2+96a^8b^4c^2-208a^3b^5c^2+9708a^5b^5c^2\\
&-96a^7b^5c^2-72a^2b^6c^2+3598a^4b^6c^2-72a^6b^6c^2+48a^5b^7c^2+24a^4b^8c^2-96ab^6c^3\\
&-30a^3b^2c^3-192a^5b^2c^3-116a^2b^3c^3+2624a^4b^3c^3-9706a^6b^3c^3-192a^8b^3c^3+1952a^3b^4c^3\\
&-9708a^5b^4c^3-800a^7b^4c^3+640a^2b^5c^3-9252a^4b^5c^3+640a^6b^5c^3-5082a^3b^6c^3-208a^5b^6c^3\\
&-448a^4b^7c^3-48a^3b^8c^3+11a^4c^4+24a^6c^4-800ab^5c^4+2a^2b^2c^4-888a^4b^2c^4+12049a^6b^2c^4\\
&+96a^8b^2c^4+1952a^3b^3c^4-9708a^5b^3c^4-800a^7b^3c^4+56b^4c^4-1136a^2b^4c^4-1747a^4b^4c^4\\
&-1136a^6b^4c^4+14336a^8b^4c^4+5082a^3b^5c^4+1952a^5b^5c^4+96b^6c^4+4399a^2b^6c^4-888a^4b^6c^4\\
&+512a^6b^6c^4-656a^3b^7c^4+7680a^5b^7c^4+24a^2b^8c^4+2816a^4b^8c^4-48a^5c^5-800ab^4c^5-208\\
&a^3b^2c^5+9708a^5b^2c^5-96a^7b^2c^5+640a^2b^3c^5-9252a^4b^3c^5+640a^6b^3c^5+5082a^3b^4c^5+\\
&1952a^5b^4c^5-192b^5c^5+5594a^2b^5c^5+2624a^4b^5c^5-29696a^6b^5c^5-192a^3b^6c^5-7680a^5b^6c^5\\
&+3584a^4b^7c^5+24a^4c^6-96ab^3c^6-72a^2b^2c^6+3598a^4b^2c^6-72a^6b^2c^6-5082a^3b^3c^6-\\
&208a^5b^3c^6+96b^4c^6+4399a^2b^4c^6-888a^4b^4c^6+512a^6b^4c^6-192a^3b^5c^6-7680a^5b^5c^6-\\
&48a^2b^6c^6+153a^4b^6c^6+48a^5b^2c^7-448a^4b^3c^7-656a^3b^4c^7+7680a^5b^4c^7+3584a^4b^5c^7\\
&+24a^4b^2c^8-48a^3b^3c^8+24a^2b^4c^8+2816a^4b^4c^8)
\end{aligned}$$

$$T_2 = (12(a-b)^2\,(a-c)^2\,(b-c)^2\,(ab-16abc^2+ac+2bc-32a^2bc-16ab^2c^2)$$

and the eigenvalues λ_i (i = 1, 2, …, 8) are

$$\lambda_3, \lambda_1 = \frac{1}{16a^2b^2c^2}\left(E_1 \pm E_2\right)$$

$$\lambda_6, \lambda_5 = \frac{1}{16a^2b^2c^2}\left(E_3 \pm E_4\right)$$

$$\lambda_8, \lambda_7 = \frac{1}{16a^2b^2c^2}\left(E_5 \pm E_6\right)$$

$$\lambda_1 = \lambda_2,\ \lambda_3 = \lambda_4 \tag{2.15}$$

where,

$$E_1 = 32a^2b^2 - 32a^2bc - 32ab^2c + 32a^2c^2 - 32abc^2 + 32b^2c^2 + 32a^4b^2c^2 - 32a^3b^3c^2 + 32a^2b^4c^2 - 32a^3b^2c^3 - 32a^2b^3c^3 + 32a^2b^2c^4$$

$$E_2 = \surd((-32a^2b^2 + 32a^2bc + 32ab^2c - 32a^2c^2 + 32abc^2 - 32b^2c^2 - 32a^4b^2c^2 + 32a^3b^3c^2 - 32a^2b^4c^2 + 32a^3b^2c^3 + 32a^2b^3c^3 - 32a^2b^2c^4)^2 - 4(768a^6b^4c^2 - 1536a^5b^5c^2 + 768a^4b^6c^2 - 1536a^6b^3c^3 + 1536a^5b^4c^3 + 1536a^4b^5c^3 - 1536a^3b^6c^3 + 768a^6b^2c^4 + 1536a^5b^3c^4 - 4608a^4b^4c^4 + 1536a^3b^5c^4 + 768a^2b^6c^4 - 1536a^5b^2c^5 + 1536a^4b^3c^5 + 1536a^3b^4c^5 - 1536a^2b^5c^5 + 768a^4b^2c^6 - 1536a^3b^3c^6 + 768a^2b^4c^6))$$

$$E_3 = 32a^2b^2 - 64ab^2c + 32a^2c^2 - 64abc^2 + 64b^2c^2 + 64a^4b^2c^2 - 64a^3b^3c^2 + 32a^2b^4c^2 - 64a^3b^2c^3 + 32a^2b^2c^4$$

$$E_4 = \surd((-32a^2b^2 + 64ab^2c - 32a^2c^2 + 64abc^2 - 64b^2c^2 - 64a^4b^2c^2 + 64a^3b^3c^2 - 32a^2b^4c^2 + 64a^3b^2c^3 - 32a^2b^2c^4)^2 - 4(1024a^6b^4c^2 - 2048a^5b^5c^2 + 1024a^4b^6c^2 - 2048a^6b^3c^3 + 2048a^5b^4c^3 + 2048a^4b^5c^3 - 2048a^3b^6c^3 + 1024a^6b^2c^4 + 2048a^5b^3c^4 - 6144a^4b^4c^4 + 2048a^3b^5c^4 + 1024a^2b^6c^4 - 2048a^5b^2c^5 + 2048a^4b^3c^5 + 2048a^3b^4c^5 - 2048a^2b^5c^5 + 1024a^4b^2c^6 - 2048a^3b^3c^6 + 1024a^2b^4c^6))$$

$$E_5 = 32a^2b^2 + 64a^2bc + 128ab^2c + 32a^2c^2 + 128abc^2 + 128b^2c^2 + 1028a^2b^2c^2 + 128a^4b^2c^2 + 128a^3b^3c^2 + 32a^2b^4c^2 + 128a^3b^2c^3 + 64a^2b^3c^3 + 32a^2b^2c^4$$

$$E_6 = \sqrt{((-32a^2b^2 - 64a^2bc - 128ab^2c - 32a^2c^2 - 128abc^2 - 128b^2c^2 - 1028a^2b^2c^2 - 128a^4b^2c^2 - 128a^3b^3c^2 - 32a^2b^4c^2 - 128a^3b^2c^3 - 64a^2b^3c^3 - 32a^2b^2c^4)^2 - 4(128a^4b^4c^2 + 256a^4b^3c^3 + 512a^3b^4c^3 - 8192a^5b^4c^3 - 4096a^4b^5c^3 + 128a^4b^2c^4 + 512a^3b^3c^4 - 8192a^5b^3c^4 + 512a^2b^4c^4 - 24576a^4b^4c^4 + 131072a^6b^4c^4 - 8192a^3b^5c^4 + 131072a^5b^5c^4 + 32768a^4b^6c^4 - 4096a^4b^3c^5 - 8192a^3b^4c^5 + 131072a^5b^4c^5 + 65536a^4b^5c^5 + 32768a^4b^4c^6))}$$

Clearly |***X′X***|, T and the eigenvalues λ_i (i = 1, 2, …, 8) are symmetric functions of a, b and c because model (2.2) is symmetrical in x_1, x_2, x_3 and x_4. In order to find D-, A- and E-optimal designs we need to find the values of a, b and c that maximize |***X′X***|, minimize T and maximize the minimum of the eigenvalues λ_i (i = 1, 2, …, 8), respectively.

If λ_0 = min (λ_i, i = 1, 2, …, 8) then from (2.15), we have λ_0 = min (λ_1, λ_5, λ_7) which gives the point at which the minimum of eigenvalues are maximized. Also, since $2a + b + c = 1$, on substituting a = (1 – (b + c)) / 2, c = 1 – (b + 2a), we obtain |***X′X***|, T and λ_i (i = 1, 2, …, 8) as functions of a and b. We have obtained values of |***X′X***|, T, λ_1, λ_5 and λ_7 for different values of a, b and c. Clearly, T, λ_1, λ_5 and λ_7 are not symmetrical in b now.

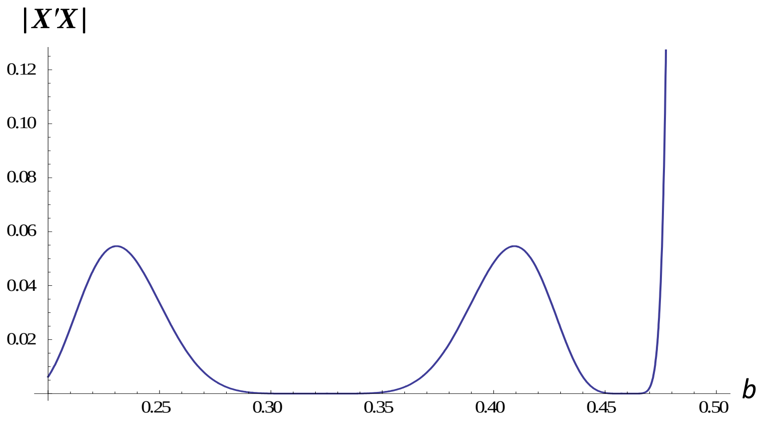


Figure 2.1. Graph of |*X′X*| against *b* for Inverse Model.

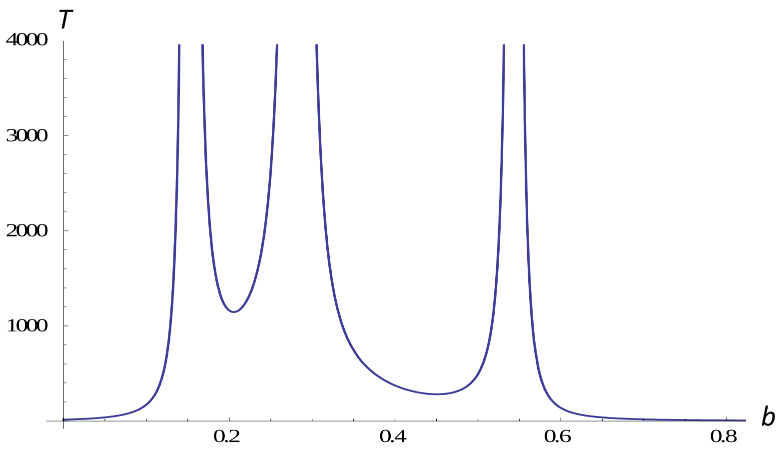


Figure 2.2. Graph of *T* against *b* for Inverse Model.

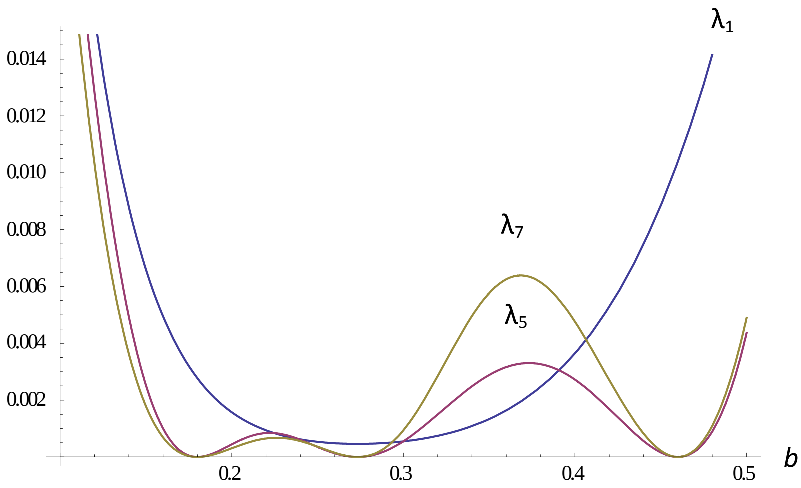


Figure 2.3. Graphs of eigenvalues λ_1, λ_5 and λ_7 against *b* for Inverse Model.

On using Mathematica with intervals of length 0.1, 0.05 and 0.01, we observe both numerically and graphically from Figure 2.1, 2.2 and 2.3, respectively that

1. $|\boldsymbol{X'X}| = 0$ when $b = 0.32$ or 0.46.
2. The curve of $|\boldsymbol{X'X}|$ is m-shaped curve. Its maximum (= 0.0546436) is attained when ($b =$ 0.2307, 0.4093).
3. T attains its minimum (= 281.229) when ($b = 0.4506$).
4. λ_0 attains its absolute maximum (= 0.0030481) at the highest point of intersection of λ_1 and λ_5 when $b = 0.3907$.

The D, A and E-optimal orthogonal block designs obtained from Design 1 for inverse model are as shown in Tables 2.1, 2.2 and 2.3, respectively.

Table 2.1. D-optimal orthogonal block Design 1 for Inverse Model

B_1				B_2			
0.1800	0.2307	0.4093	0.1800	0.1800	0.1800	0.4093	0.2307
0.2307	0.4093	0.1800	0.1800	0.2307	0.1800	0.1800	0.4093
0.4093	0.1800	0.1800	0.2307	0.4093	0.2307	0.1800	0.1800
0.1800	0.1800	0.2307	0.4093	0.1800	0.4093	0.2307	0.1800
0.1800	0.4093	0.1800	0.2307	0.1800	0.4093	0.2307	0.1800
0.2307	0.1800	0.1800	0.4093	0.2307	0.1800	0.4093	0.1800
0.4093	0.1800	0.2307	0.1800	0.4093	0.1800	0.1800	0.2307
0.1800	0.2307	0.4093	0.1800	0.1800	0.2307	0.1800	0.4093
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.2. A-optimal orthogonal block Design 1 for Inverse Model

B_1				B_2			
0.1987	0.4506	0.1520	0.1987	0.1987	0.1987	0.1520	0.4506
0.4506	0.1520	0.1987	0.1987	0.4506	0.1987	0.1987	0.1520
0.1520	0.1987	0.1987	0.4506	0.1520	0.4506	0.1987	0.1987
0.1987	0.1987	0.4506	0.1520	0.1987	0.1520	0.4506	0.1987
0.1987	0.1520	0.1987	0.4506	0.1987	0.1520	0.4506	0.1987
0.4506	0.1987	0.1987	0.1520	0.4506	0.1987	0.1520	0.1987
0.1520	0.1987	0.4506	0.1987	0.1520	0.1987	0.1987	0.4506
0.1987	0.4506	0.1520	0.1987	0.1987	0.4506	0.1987	0.1520
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.3. E-optimal orthogonal block Design 1 for Inverse Model

B_1				B_2			
0.2146	0.3907	0.1800	0.2146	0.2146	0.2146	0.1800	0.3907
0.3907	0.1800	0.2146	0.2146	0.3907	0.2146	0.2146	0.1800
0.1800	0.2146	0.2146	0.3907	0.1800	0.3907	0.2146	0.2146
0.2146	0.2146	0.3907	0.1800	0.2146	0.1800	0.3907	0.2146
0.2146	0.1800	0.2146	0.3907	0.2146	0.1800	0.3907	0.2146
0.3907	0.2146	0.2146	0.1800	0.3907	0.2146	0.1800	0.2146
0.1800	0.2146	0.3907	0.2146	0.1800	0.2146	0.2146	0.3907
0.2146	0.3907	0.1800	0.2146	0.2146	0.3907	0.2146	0.1800
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal orthogonal block designs obtained from Design 2 for inverse model are as shown in Tables 2.4, 2.5 and 2.6, respectively.

Table 2.4. D-optimal orthogonal block Design 2 for Inverse Model

B_1				B_2			
0.1800	0.2307	0.4093	0.1800	0.1800	0.1800	0.4093	0.2307
0.2307	0.4093	0.1800	0.1800	0.2307	0.1800	0.1800	0.4093
0.4093	0.1800	0.1800	0.2307	0.4093	0.2307	0.1800	0.1800
0.1800	0.1800	0.2307	0.4093	0.1800	0.4093	0.2307	0.1800
0.1800	0.1800	0.2307	0.4093	0.1800	0.2307	0.1800	0.4093
0.2307	0.1800	0.4093	0.1800	0.2307	0.4093	0.1800	0.1800
0.4093	0.2307	0.1800	0.1800	0.4093	0.1800	0.2307	0.1800
0.1800	0.4093	0.1800	0.2307	0.1800	0.1800	0.4093	0.2307
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.5. A-optimal orthogonal block Design 2 for Inverse Model

B_1				B_2			
0.1987	0.4506	0.1520	0.1987	0.1987	0.1987	0.1520	0.4506
0.4506	0.1520	0.1987	0.1987	0.4506	0.1987	0.1987	0.1520
0.1520	0.1987	0.1987	0.4506	0.1520	0.4506	0.1987	0.1987
0.1987	0.1987	0.4506	0.1520	0.1987	0.1520	0.4506	0.1987
0.1987	0.1987	0.4506	0.1520	0.1987	0.4506	0.1987	0.1520
0.4506	0.1987	0.1520	0.1987	0.4506	0.1520	0.1987	0.1987
0.1520	0.4506	0.1987	0.1987	0.1520	0.1987	0.4506	0.1987
0.1987	0.1520	0.1987	0.4506	0.1987	0.1987	0.1520	0.4506
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.6. E-optimal orthogonal block Design 2 for Inverse Model

B_1				B_2			
0.2146	0.3907	0.1800	0.2146	0.2146	0.2146	0.1800	0.3907
0.3907	0.1800	0.2146	0.2146	0.3907	0.2146	0.2146	0.1800
0.1800	0.2146	0.2146	0.3907	0.1800	0.3907	0.2146	0.2146
0.2146	0.2146	0.3907	0.1800	0.2146	0.1800	0.3907	0.2146
0.2146	0.2146	0.3907	0.1800	0.2146	0.3907	0.2146	0.1800
0.3907	0.2146	0.1800	0.2146	0.3907	0.1800	0.2146	0.2146
0.1800	0.3907	0.2146	0.2146	0.1800	0.2146	0.3907	0.2146
0.2146	0.1800	0.2146	0.3907	0.2146	0.2146	0.1800	0.3907
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal orthogonal block designs obtained from Design 3 for inverse model are as shown in Tables 2.7, 2.8 and 2.9, respectively.

Table 2.7. D-optimal orthogonal block Design 3 for Inverse Model

B_1				B_2			
0.1800	0.4093	0.1800	0.2307	0.1800	0.4093	0.2307	0.1800
0.2307	0.1800	0.1800	0.4093	0.2307	0.1800	0.4093	0.1800
0.4093	0.1800	0.2307	0.1800	0.4093	0.1800	0.1800	0.2307
0.1800	0.2307	0.4093	0.1800	0.1800	0.2307	0.1800	0.4093
0.1800	0.1800	0.2307	0.4093	0.1800	0.2307	0.1800	0.4093
0.2307	0.1800	0.4093	0.1800	0.2307	0.4093	0.1800	0.1800
0.4093	0.2307	0.1800	0.1800	0.4093	0.1800	0.2307	0.1800
0.1800	0.4093	0.1800	0.2307	0.1800	0.1800	0.4093	0.2307
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.8. A-optimal orthogonal block Design 3 for Inverse Model

B_1				B_2			
0.1987	0.1520	0.1987	0.4506	0.1987	0.1520	0.4506	0.1987
0.4506	0.1987	0.1987	0.1520	0.4506	0.1987	0.1520	0.1987
0.1520	0.1987	0.4506	0.1987	0.1520	0.1987	0.1987	0.4506
0.1987	0.4506	0.1520	0.1987	0.1987	0.4506	0.1987	0.1520
0.1987	0.1987	0.4506	0.1520	0.1987	0.4506	0.1987	0.1520
0.4506	0.1987	0.1520	0.1987	0.4506	0.1520	0.1987	0.1987
0.1520	0.4506	0.1987	0.1987	0.1520	0.1987	0.4506	0.1987
0.1987	0.1520	0.1987	0.4506	0.1987	0.1987	0.1520	0.4506
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.9. E-optimal orthogonal block Design 3 for Inverse Model

B_1				B_2			
0.2146	0.1800	0.2146	0.3907	0.2146	0.1800	0.3907	0.2146
0.3907	0.2146	0.2146	0.1800	0.3907	0.2146	0.1800	0.2146
0.1800	0.2146	0.3907	0.2146	0.1800	0.2146	0.2146	0.3907
0.2146	0.3907	0.1800	0.2146	0.2146	0.3907	0.2146	0.1800
0.2146	0.2146	0.3907	0.1800	0.2146	0.3907	0.2146	0.1800
0.3907	0.2146	0.1800	0.2146	0.3907	0.1800	0.2146	0.2146
0.1800	0.3907	0.2146	0.2146	0.1800	0.2146	0.3907	0.2146
0.2146	0.1800	0.2146	0.3907	0.2146	0.2146	0.1800	0.3907
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 2.10 depicts the values of parameters *a*, *b* and *c* for Draper and St. John's (1977) quadratic mixture model with inverse terms in four components.

Table 2.10. The numerical values of the design parameters for four component mixtures based on F-squares

	a	*b*	*c*
D – optimality	0.1800	0.2307, 0.4093	0.4093, 0.2307
A – optimality	0.1987	0.4506	0.1520
E – optimality	0.2146	0.3907	0.1800

2.5. Conclusions

In this chapter, we have constructed optimal orthogonal designs in two blocks based on F-squares for Draper and St. John's (1977) quadratic mixture model with inverse terms in four components when two component proportions are at the same level. In Section 2.2, we have obtained conditions for the orthogonal blocking of blends for Draper and St. John's (1977) model. From the results in Section 2.4, we find that Design 1, Design 2 and Design 3 are D-optimal when $a = 0.18$, $c = 0.64 - b$ where $b = 0.2307, 0.4093$; A-optimal when $c = 0.1520$, $a = 0.424 - b/2$, where $b = 0.4506$ and E-optimal when $c = 0.18$, $a = 0.41 - b/2$, where $b = 0.3907$, respectively. Moreover, since the function $|\boldsymbol{X'X}|$ is symmetrical in b and c, the D-optimality of the designs considered, viz., Design 1, Design 2 and Design 3 is maintained by the interchange of b and c.

Chapter 3

F-SQUARES BASED OPTIMAL DESIGNS FOR REDUCED CUBIC CANONICAL MODEL IN FOUR MIXTURE COMPONENTS

3.1. Introduction

In the previous chapter, we have obtained D-, A- and E-optimal orthogonal block designs in two blocks for Draper and St. John's (1977) inverse model for $q = 4$. In this chapter, we will construct F-square based D-, A- and E-optimal block designs in four components for the reduced cubic canonical model.

The full cubic canonical polynomial due to Scheffé (1958) is as given by

$$E(y)=\sum_{i=1}^{q}\beta_i x_i + \sum_{1\le i<j\le q}\beta_{ij}x_i x_j + \sum_{1\le i<j\le q}\beta_{ij}x_i x_j(x_i - x_j) + \sum_{1\le i<j<k\le q}\beta_{ijk}x_i x_j x_k. \qquad (3.1)$$

We present the following reduced cubic canonical mixture model. This new model is beneficial in determining synergism, if any amongst the mixture components.

$$E(y)=\sum_{i=1}^{q}\beta_i x_i + \sum_{1\le i<j\le q}\beta_{ij}x_i x_j(x_i - x_j). \qquad (3.2)$$

The full cubic canonical polynomial model (3.1) has $q\ (q + 1)\ (q + 2)/6$ terms whereas the reduced cubic canonical mixture model (3.2) has $q\ (q + 1) / 2$ terms. The reduced cubic canonical mixture model (3.2) enables us to experiment with fewer number of observations, viz; $q\ (q^2 – 1) / 6$ less observations as compared to the full cubic model.

In Section 3.2, we will discuss the orthogonality conditions for the reduced cubic canonical model. In Section 3.3, we will present optimal blocked four component mixture designs. These designs are useful when the mixture experiment does not involve any extraneous factors affecting the response of interest. In Section 3.4, we will present conditions for the orthogonal blocking of blends for the reduced cubic canonical model and obtain the D-, A- and E-optimalities of these designs for the situations when the response is dependent on factors other than the mixture variables. These classes of designs are based on F-squares as given in Aggarwal et al. (2009).

3.2. Blocking Conditions

In case of two blocks, say B_1 and B_2 having m_1 and m_2 blends such that $m_1 + m_2 = m$ mixture blends, the model (3.3), with block effect γ is

$$E(y)=\sum_{i=1}^{q}\beta_i x_{iu} + \sum_{1\le i<j\le q}\beta_{ij}x_{iu}x_{ju}(x_{iu}-x_{ju})+\gamma Z_u\,; \quad u=1,2,...,m. \tag{3.3}$$

Here $Z_u = -1$, for the blends in block B_1, and $Z_u = 1$, for the blends in block B_2 and e_u's are random errors which are independently distributed with mean 0 and same variance σ^2. Model (3.3) does not contain the product terms of x_i and Z_u. In matrix notation, model (3.3) may be expressed as follows

$$E(y) = \boldsymbol{X}\beta + \boldsymbol{Z\gamma} \tag{3.4}$$

where $\boldsymbol{X}$ is the $m \times (q + q\,(q\text{-}1) / 2)$ matrix corresponding to the mixture part, β is the $(q + q\,(q\text{-}1) / 2) \times 1$ column vector of unknown parameters, $\boldsymbol{\gamma}$ is the block effect parameter and Z is the $m \times 1$ column vector corresponding to the block variable $\boldsymbol{Z}$. For the reduced cubic canonical model, we have obtained the following conditions required for the orthogonal blocking of mixture blends in the presence of process variables.

$$\begin{aligned}&\sum_{u=1}^{m_w} x_{iu} = k_i\,, && i = 1,\ 2,\ldots,q\\ &\sum_{u=1}^{m_w} x_{iu}x_{ju}(x_{iu}-x_{ju}) = k'_{ij} && \forall\, w = 1,\ 2;\ \ i = 1,\ 2,\ldots,q\end{aligned} \tag{3.5}$$

where k_i's and k'_{ij}'s are constants.

3.3. Blocked Four Component Mixture Designs with Reduced Cubic Canonical Terms using F-squares

For four component mixtures, eleven distinct runs are required to estimate all the parameters in (3.3). With a single block variable at two levels, $Z = -1$ and $Z = +1$, we take one block at $Z = -1$ and the other block at $Z = +1$. We consider the designs viz, Design 1, Design 2 and Design 3 given as (2.7), (2.8) and (2.9), respectively in Chapter 2. These designs are given by Aggarwal et al. (2009).

In this chapter, we use this class of designs to obtain D-, A- and E-optimal block designs for the different cases depending on whether the process variables are absent or present as shown in Section 3.4 and 3.5, respectively.

3.4. Optimal Blocked Four Component Mixture Designs

In this section, we derive optimal blocked four component mixture designs for the reduced cubic canonical model for the situation when the orthogonality conditions given in (3.5) are not satisfied. This situation may arise when no process variables are present in the setup of the mixture experiments to affect the response of interest. Lemonade is made by mixing lemon juice, salt, sugar and black salt in different proportions and the response of interest is the tangy flavour of the lemonade. Here the response is independent of the levels of factors other than the mixture ingredients.

The form of matrix $\boldsymbol{X}$ for Design 1 for the model (3.2) is as given in (3.6).

$$\boldsymbol{X}=\begin{bmatrix}
a & b & c & a & ab(a-b) & ac(a-c) & 0 & bc(b-c) & ba(b-a) & ca(c-a)\\
b & c & a & a & bc(b-c) & ba(b-a) & ba(b-a) & ca(c-a) & ca(c-a) & 0\\
c & a & a & b & ca(c-a) & ca(c-a) & cb(c-b) & 0 & ab(a-b) & ab(a-b)\\
a & a & b & c & 0 & ab(a-b) & ac(a-c) & ab(a-b) & ac(a-c) & bc(b-c)\\
a & c & a & b & ac(a-c) & 0 & ab(a-b) & ca(c-a) & cb(c-b) & ab(a-b)\\
b & a & a & c & ba(b-a) & ba(b-a) & bc(b-c) & 0 & ac(a-c) & ac(a-c)\\
c & a & b & a & ca(c-a) & cb(c-b) & ca(c-a) & ab(a-b) & 0 & ba(b-a)\\
a & b & c & a & ab(a-b) & ac(a-c) & 0 & bc(b-c) & ba(b-a) & ca(c-a)\\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0\\
a & a & c & b & 0 & ac(a-c) & ab(a-b) & ac(a-c) & ab(a-b) & cb(c-b)\\
b & a & a & c & ba(b-a) & ba(b-a) & bc(b-c) & 0 & ac(a-c) & ac(a-c)\\
c & b & a & a & cb(c-b) & ca(c-a) & ca(c-a) & ba(b-a) & ba(b-a) & 0\\
a & c & b & a & ac(a-c) & ab(a-b) & 0 & cb(c-b) & ca(c-a) & ba(b-a)\\
a & c & b & a & ac(a-c) & ab(a-b) & 0 & cb(c-b) & ca(c-a) & ba(b-a)\\
b & a & c & a & ba(b-a) & bc(b-c) & ba(b-a) & ac(a-c) & 0 & ca(c-a)\\
c & a & a & b & ca(c-a) & ca(c-a) & cb(c-b) & 0 & ab(a-b) & ab(a-b)\\
a & b & a & c & ab(a-b) & 0 & ac(a-c) & ba(b-a) & bc(b-c) & ac(a-c)\\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix} \tag{3.6}$$

The forms of $\boldsymbol{X'X}$ for Design 1, Design 2 and Design 3 are as given in (3.7), (3.8) and (3.9), respectively.

$$\boldsymbol{X'X} = \left|\begin{array}{cccc|cccccc} P & R & R & Q & S & S & T & O & U & U \\ R & P & Q & R & V & U & O & T & S & W \\ R & Q & P & R & U & V & O & X & W & S \\ Q & R & R & P & W & W & X & O & V & V \\ \hline S & V & U & W & A & B & C & D & E & F \\ S & U & V & W & B & A & C & C & F & E \\ T & O & O & X & C & C & G & O & C & C \\ O & T & X & O & D & C & O & G & C & D \\ U & S & W & V & E & F & C & C & A & B \\ U & W & S & V & F & E & C & D & B & A \end{array}\right| \tag{3.7}$$

$$\boldsymbol{X'X} = \left|\begin{array}{cccc|cccccc} P & Q & R & R & T & S & S & W & W & O \\ Q & P & R & R & X & W & W & S & S & O \\ R & R & P & Q & O & V & U & V & U & T \\ R & R & Q & P & O & U & V & U & V & X \\ \hline T & X & O & O & G & C & C & D & D & O \\ S & W & V & U & C & A & B & B & H & D \\ S & W & U & V & C & B & A & H & B & C \\ W & S & V & U & D & B & H & A & B & D \\ W & S & U & V & D & H & B & B & A & C \\ O & O & T & X & O & D & C & D & C & G \end{array}\right| \tag{3.8}$$

$$\boldsymbol{X'X} = \left|\begin{array}{cccc|cccccc} P & R & Q & R & S & T & S & U & O & W \\ R & P & R & Q & V & O & U & S & T & U \\ Q & R & P & R & W & X & W & V & O & S \\ R & Q & R & P & U & O & V & W & X & V \\ \hline S & V & W & U & A & C & B & E & D & H \\ T & O & X & O & C & G & C & C & O & D \\ S & U & W & V & B & C & A & F & C & B \\ U & S & V & W & E & C & F & A & C & E \\ O & T & O & X & D & O & C & C & G & C \\ W & U & S & V & H & D & B & E & C & A \end{array}\right| \tag{3.9}$$

where,

$$P=\frac{1}{8}+8a^2+4b^2+4c^2$$

$$Q=\frac{1}{8}+4a^2+4ab+4ac+4bc$$

$$R=\frac{1}{8}+2a^2+6ab+6ac+2bc$$

$$O=0$$

$$S=3a^2(a-b)b-3a(a-b)b^2+3a^2(a-c)c+b^2(b-c)c-3a(a-c)c^2-b(b-c)c^2$$

$$T=2a^2(a-b)b-2a(a-b)b^2+2a^2(a-c)c+2b^2(b-c)c-2a(a-c)c^2-2b(b-c)c^2$$

$$U=a^2(b-a)b+a(a-b)bc-a^2(a-c)c+ab(a-c)c$$

$$V=3a^2(b-a)b+3a(a-b)b^2-3a^2(a-c)c-b^2(b-c)c+3a(a-c)c^2+b(b-c)c^2$$

$$W=a^2(a-b)b-a(a-b)bc+a^2(a-c)c-ab(a-c)c$$

$$X=2a(a-c)c^2+2b(b-c)c^2-2a^2(a-b)b+2a(a-b)b^2-2a^2(a-c)c-2b^2(b-c)c$$

$$A=6a^2(a-b)^2b^2+6a^2(a-c)^2c^2+2b^2(b-c)^2c^2$$

$$B=2a^2(a-b)^2b^2+4a^2(a-b)b(a-c)c-2a(a-b)b^2(b-c)c+2a^2(a-c)^2c^2+2ab(a-c)(b-c)c^2$$

$$C=a^2(a-b)^2b^2+2a^2(a-b)b(a-c)c-3a(a-b)b^2(b-c)c+a^2(a-c)^2c^2+3ab(a-c)(b-c)c^2$$

$$D=3a(a-b)b^2(b-c)c-a^2(a-b)^2b^2-2a^2(a-b)b(a-c)c-a^2(a-c)^2c^2-3ab(a-c)(b-c)c^2$$

$$E=2a(a-b)b^2(b-c)c-2a^2(a-b)^2b^2-4a^2(a-b)b(a-c)c-2a^2(a-c)^2c^2-2ab(a-c)(b-c)c^2$$

$$F=4a^2(b-a)b(a-c)c$$

$$G=4a^2(a-b)^2b^2+4a^2(a-c)^2c^2+4b^2(b-c)^2c^2$$

$$H=4a^2(a-b)b(a-c)c \qquad (3.10)$$

Clearly, we have different forms of ***X′X*** for the three designs, viz., Design 1, Design 2 and Design 3. However same forms of $|\boldsymbol{X'X}|$, T = Trace $(\boldsymbol{X'X})^{-1}$ and the eigenvalues λ_i (i = 1, 2, …, 10) of $\boldsymbol{X'X}$ are obtained.

$|\boldsymbol{X'X}|$

$$=256(a-b)^{12}(a-c)^{12}(b-c)^{12}(2a+b+c)^{6}(1+32a^2+32ab+8b^2+32ac+16bc+8c^2) \tag{3.11}$$

$$T = T_1 / T_2 \tag{3.12}$$

where,

$$\begin{aligned}
T_1 = {} & 20a^2 + 640a^4 + 640a^3b + 10b^2 + 480a^2b^2 + 15a^4b^2 + 496a^6b^2 + 320ab^3 \\
& - 30a^3b^3 - 496a^5b^3 + 80b^4 + 15a^2b^4 - 372a^4b^4 + 248a^3b^5 \\
& + 124a^2b^6 + 640a^3c + 320a^2bc + 10a^4bc + 288a^6bc + 320ab^2c \\
& - 10a^3b^2c + 496a^5b^2c + 160b^3c - 10a^2b^3c - 1248a^4b^3c + 10ab^4c \\
& - 72a^3b^4c + 464a^2b^5c + 72ab^6c + 10c^2 + 480a^2c^2 + 15a^4c^2 \\
& + 496a^6c^2 + 320abc^2 - 10a^3bc^2 + 496a^5bc^2 + 160b^2c^2 + 30a^2b^2c^2 \\
& + 680a^4b^2c^2 - 10ab^3c^2 - 176a^3b^3c^2 + 5b^4c^2 + 372a^2b^4c^2 \\
& + 248ab^5c^2 + 44b^6c^2 + 320ac^3 - 30a^3c^3 - 496a^5c^3 + 160bc^3 \\
& - 10a^2bc^3 - 1248a^4bc^3 - 10ab^2c^3 - 176a^3b^2c^3 - 10b^3c^3 \\
& - 640a^2b^3c^3 - 320ab^4c^3 + 80c^4 + 15a^2c^4 - 372a^4c^4 + 10abc^4 \\
& - 72a^3bc^4 + 5b^2c^4 + 372a^2b^2c^4 - 320ab^3c^4 - 88b^4c^4 + 248a^3c^5 \\
& + 464a^2bc^5 + 248ab^2c^5 + 124a^2c^6 + 72abc^6 + 44b^2c^6
\end{aligned}$$

$$T_2 = 2(a-b)^2(a-c)^2(b-c)^2(2a+b+c)^2(1+32a^2+32ab+8b^2+32ac+16bc+8c^2)$$

The expressions of eigenvalues λ_i (i = 1, 2, …, 10) are very lengthy and are available in the attached C.D. On using Mathematica with intervals of length 0.1, 0.01 and 0.05, we conclude that the D-, A- and E-optimal values of the three designs are obtained at the point a = 0. The forms of $\boldsymbol{X'X}$ for Design 1, Design 2 and Design 3 in this case is as given in (3.7), (3.8) and (3.9), respectively with

$$P = \frac{1}{8} + 4b^2 + 4c^2$$

$$Q = \frac{1}{8} + 4bc$$

$$R = \frac{1}{8} + 2bc$$

$$S = b^2(b-c)c - b(b-c)c^2$$

$$T = 2b^2(b-c)c - 2b(b-c)c^2$$

$$V = b(b-c)c^2 - b^2(b-c)c$$

$$X = 2b(b-c)c^2 - 2b^2(b-c)c$$

$$A = 2b^2(b-c)^2 c^2$$

$$G = 4b^2(b-c)^2 c^2$$

$$U = W = B = C = D = E = F = H = 0 \tag{3.13}$$

For all that three designs, we have obtained the following results.

$$|\boldsymbol{X'X}| = 256b^{12}(b-c)^{12}c^{12}(b+c)^6(1+8b^2+16bc+8c^2) \tag{3.14}$$

$$T = T_1 / T_2 \tag{3.15}$$

where,

$$T_1 = 10b^2 + 80b^4 + 160b^3c + 10c^2 + 160b^2c^2 + 5b^4c^2 + 44b^6c^2 + 160bc^3 - 10b^3c^3 + 80c^4 + 5b^2c^4 - 88b^4c^4 + 44b^2c^4$$

$$T_2 = 2b^2(b-c)^2c^2(b+c)^2(1+8b^2+16bc+8c^2)$$

The expression of eigenvalues λ_i (i = 1, 2, …, 10) are very lengthy and are available in the attached C.D.

Since the model (3.2) is symmetrical in x_1, x_2, x_3 and x_4, $|\boldsymbol{X'X}|$, T and the eigenvalues λ_i (i = 1, 2, …, 10) are symmetric functions of b and c. In order to find the D-, A- and

E-optimal designs for the reduced cubic canonical model, we need to find the values of b and c that maximize $|\boldsymbol{X'X}|$, minimize T and maximize the minimum of the eigenvalues λ_i (i = 1, 2, …, 10), respectively. If $\lambda_0 = \min(\lambda_i, i = 1, 2, \ldots, 10)$, we have $\lambda_0 = \min(\lambda_3, \lambda_5)$. Also since $b + c = 1$, on substituting $c = 1 - b$, we obtain $|\boldsymbol{X'X}|$, T and λ_i (i = 1, 2, …, 10) as functions of b alone. We have obtained different values of $|\boldsymbol{X'X}|$, T, λ_3 and λ_5 for $b \in [0, 1]$.

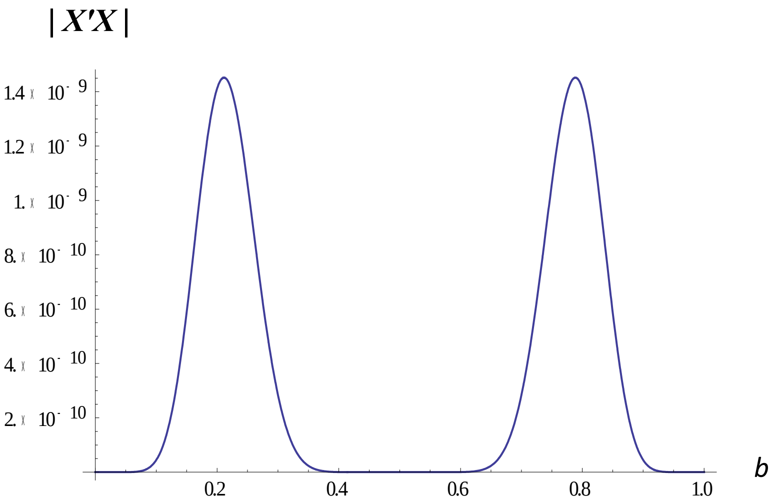


Figure 3.1. Graph of $|X'X|$ against b for the Reduced Cubic Canonical Model when a = 0.

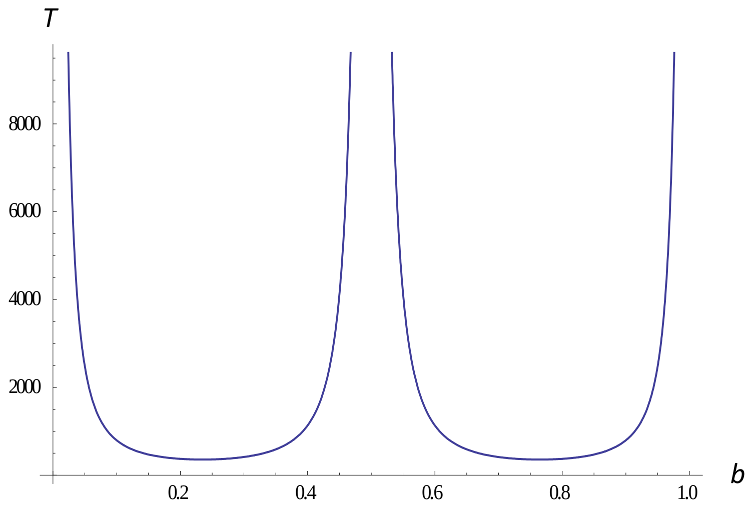


Figure 3.2. Graph of T against b for the Reduced Cubic Canonical Model when a = 0.

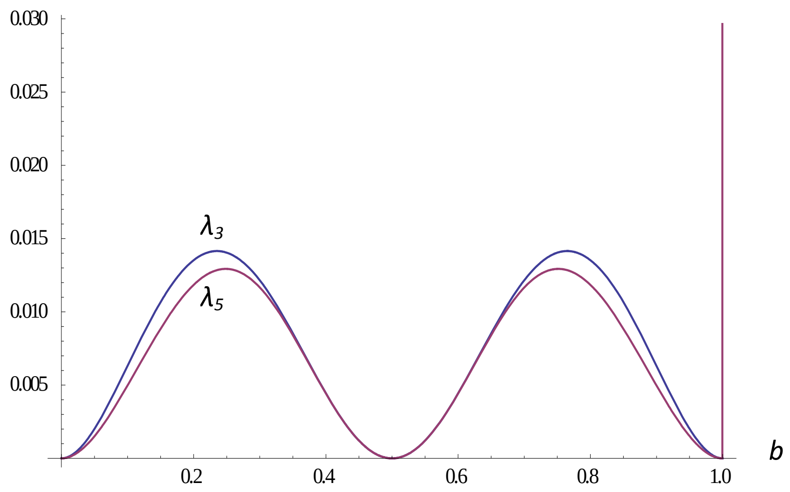


Figure 3.3. Graphs of eigenvalues λ_3 and λ_5 against b for the Reduced Cubic Canonical Model when a = 0.

Figure 3.1, 3.2 and 3.3 represent the graphs of $|\boldsymbol{X'X}|$, T and eigenvalues λ_3 and λ_5 against b for the Reduced Cubic Canonical Model when $a = 0$.

We observe both numerically and graphically that

1. $|\boldsymbol{X'X}| = 0$ when $b = 0$, $\frac{1}{2}$ or 1.
2. The curve of $|\boldsymbol{X'X}|$ is an m-shaped curve. Its maximum ($= 1.4 \times 10^{-9}$) is attained when $b = 0.205902, 0.784231$.
3. T attains its minimum (= 355.454) when $b = 0.235058, 0.764942$.
4. $\lambda_0 = \min(\lambda_3, \lambda_5)$ attains its absolute maximum (= 0.009576) at $b = 0.346712$, 0.657281.

Since all the three designs, viz; Design 1, Design 2 and Design 3 are symmetric in b and c, we get similar results for both the cases $b = 0$ and $c = 0$. We have considered the case $c = 0$ to accommodate for the situation in which an experimenter wishes to include at most ternary blends (excluding the centroid) in the mixture model. The forms of $\boldsymbol{X'X}$ for Design 1, Design 2 and Design 3 in this case are as given in (3.7), (3.8) and (3.9), respectively with the following modifications.

$$P = \frac{1}{8} + 8a^2 + 4b^2$$

$$Q = \frac{1}{8} + 4a^2 + 4ab$$

$R = \frac{1}{8} + 2a^2 + 6ab$

$S = 3a^2(a-b)b - 3a(a-b)b^2$

$T = 2a^2(a-b)b - 2a(a-b)b^2$

$U = a^2(b-a)b$

$V = 3a(a-b)b^2 - 3a^2(a-b)b$

$W = a^2(a-b)b$

$X = 2a(a-b)b^2 - 2a^2(a-b)b$

$A = 6a^2(a-b)^2 b^2$

$B = 2a^2(a-b)^2 b^2$

$C = a^2(a-b)^2 b^2$

$D = -a^2(a-b)^2 b^2$

$E = -2a^2(a-b)^2 b^2$

$F = H = 0$

$G = 4a^2(a-b)^2 b^2$ (3.16)

For all the three designs, we have obtained the following results.

$|\boldsymbol{X'X}| = 256a^{12}(a-b)^{12}b^{12}(2a+b)^6(1+32a^2+32ab+8b^2)$ (3.17)

$T = T_1 / T_2$ (3.18)

where,

$$T_1 = 20a^2 + 640a^4 + 640a^3b + 10b^2 + 480a^2b^2 + 15a^4b^2 + 496a^6b^2 + 320ab^3 - 30a^3b^3 - 496a^5b^3 + 80b^4 + 15a^2b^4 - 372a^4b^4 + 248a^3b^5 + 124a^2b^6$$

$$T_2 = 2a^2(a-b)^2b^2(2a+b)^2(1+32a^2+32ab+8b^2)$$

The expressions for eigenvalues λ_i (i = 1, 2, …, 10) are very lengthy and are available in the attached C.D.

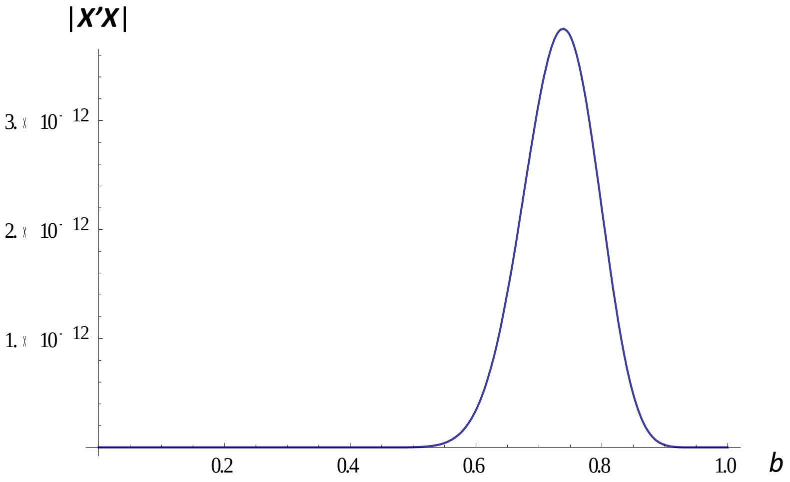


Figure 3.4. Graph of |*X′X*| against *b* for the Reduced Cubic Canonical Model when *c* = 0.

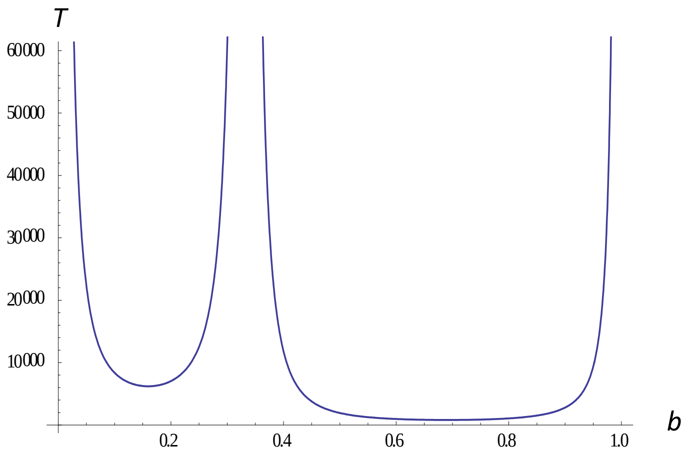


Figure 3.5. Graph of *T* against *b* for the Reduced Cubic Canonical Model when *c* = 0.

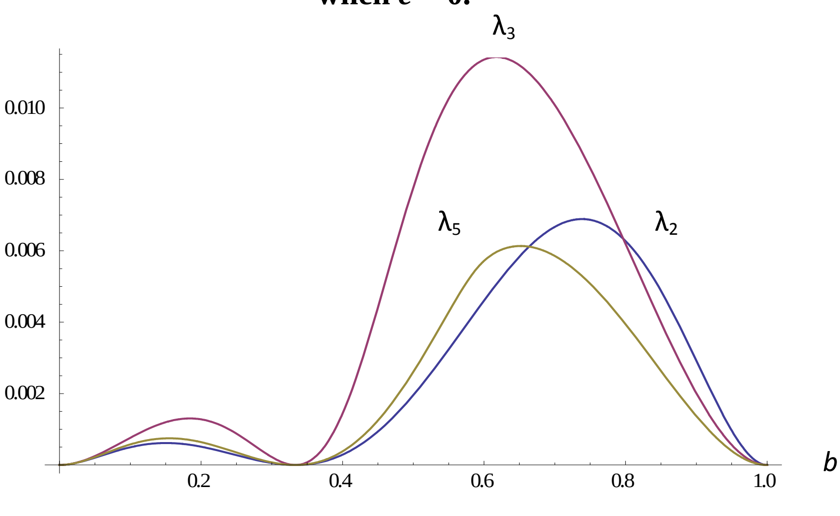


Figure 3.6. Graphs of eigenvalues λ_2, λ_3 and λ_5 against *b* for the Reduced Cubic Canonical Model when *c* = 0.

Figure 3.4, 3.5 and 3.6 represent the graphs of $|\boldsymbol{X'X}|$, T and eigenvalues λ_2, λ_3 and λ_5 against b for the Reduced Cubic Canonical Model when $c = 0$.

We observe both numerically and graphically that

1. $|\boldsymbol{X'X}| = 0$ when $0 \le b \le 1/2$ or $b = 1$.
2. The curve of $|\boldsymbol{X'X}|$ is an inverted V-shaped curve. Its maximum ($= 3.8431 \times 10^{-12}$) is attained when $b = 0.73856$.
3. T attains its minimum (= 808.212) when $b = 0.690308$.
4. $\lambda_0 = \min(\lambda_2, \lambda_3, \lambda_5)$ attains its absolute maximum (= 0.00613316) at $b = 0.651645$.

The D, A and E-optimal block designs obtained from Design 1 for the Reduced Cubic Canonical Model are as shown in Tables 3.2, 3.3 and 3.4, respectively.

Table 3.2. D-optimal blocked Design 1 when *a* = 0

$\mathbf{B_1}$				$\mathbf{B_2}$			
0	0.2059	0.7842	0	0	0	0.7842	0.2059
0.2059	0.7842	0	0	0.2059	0	0	0.7842
0.7842	0	0	0.2059	0.7842	0.2059	0	0
0	0	0.2059	0.7842	0	0.7842	0.2059	0
0	0.7842	0	0.2059	0	0.7842	0.2059	0
0.2059	0	0	0.7842	0.2059	0	0.7842	0
0.7842	0	0.2059	0	0.7842	0	0	0.2059
0	0.2059	0.7842	0	0	0.2059	0	0.7842
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.3. A-optimal blocked Design 1 when *a* = 0

$\mathbf{B_1}$				$\mathbf{B_2}$			
0	0.235058	0.764942	0	0	0	0.764942	0.235058
0.235058	0.764942	0	0	0.235058	0	0	0.764942
0.764942	0	0	0.235058	0.764942	0.235058	0	0
0	0	0.235058	0.764942	0	0.764942	0.235058	0
0	0.764942	0	0.235058	0	0.764942	0.235058	0
0.235058	0	0	0.764942	0.235058	0	0.764942	0
0.764942	0	0.235058	0	0.764942	0	0	0.235058
0	0.235058	0.764942	0	0	0.235058	0	0.764942
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.4. E-optimal blocked Design 1 when *a* = 0

B1				B2			
0	0.346712	0.657281	0	0	0	0.657281	0.346712
0.346712	0.657281	0	0	0.346712	0	0	0.657281
0.657281	0	0	0.346712	0.657281	0.346712	0	0
0	0	0.346712	0.657281	0	0.657281	0.346712	0
0	0.657281	0	0.346712	0	0.657281	0.346712	0
0.346712	0	0	0.657281	0.346712	0	0.657281	0
0.657281	0	0.346712	0	0.657281	0	0	0.346712
0	0.346712	0.657281	0	0	0.346712	0	0.657281
1/4	¼	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal block designs obtained from Design 2 for the Reduced Cubic Canonical Model are as shown in Tables 3.5, 3.6 and 3.7, respectively.

Table 3.5. D-optimal blocked Design 2 when *a* = 0

B1				B2			
0	0.2059	0.7842	0	0	0	0.7842	0.2059
0.2059	0.7842	0	0	0.2059	0	0	0.7842
0.7842	0	0	0.2059	0.7842	0.2059	0	0
0	0	0.2059	0.7842	0	0.7842	0.2059	0
0	0	0.2059	0.7842	0	0.2059	0	0.7842
0.2059	0	0.7842	0	0.2059	0.7842	0	0
0.7842	0.2059	0	0	0.7842	0	0.2059	0
0	0.7842	0	0.2059	0	0	0.7842	0.2059
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.6. A-optimal blocked Design 2 when *a* = 0

B1				B2			
0	0.235058	0.764942	0	0	0	0.764942	0.235058
0.235058	0.764942	0	0	0.235058	0	0	0.764942
0.764942	0	0	0.235058	0.764942	0.235058	0	0
0	0	0.235058	0.764942	0	0.764942	0.235058	0
0	0	0.235058	0.764942	0	0.235058	0	0.764942
0.235058	0	0.764942	0	0.235058	0.764942	0	0
0.764942	0.235058	0	0	0.764942	0	0.235058	0
0	0.764942	0	0.235058	0	0	0.764942	0.235058
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.7. E-optimal blocked Design 2 when *a* = 0

B1				B2			
0	0.346712	0.657281	0	0	0	0.657281	0.346712
0.346712	0.657281	0	0	0.346712	0	0	0.657281
0.657281	0	0	0.346712	0.657281	0.346712	0	0
0	0	0.346712	0.657281	0	0.657281	0.346712	0
0	0	0.346712	0.657281	0	0.346712	0	0.657281
0.346712	0	0.657281	0	0.346712	0.657281	0	0
0.657281	0.346712	0	0	0.657281	0	0.346712	0
0	0.657281	0	0.346712	0	0	0.657281	0.346712
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal block designs obtained from Design 3 for the Reduced Cubic Canonical Model are as shown in Tables 3.8, 3.9 and 3.10, respectively.

Table 3.8. D-optimal of optimal blocked Design 3 when *a* = 0

B1				B2			
0	0.7842	0	0.2059	0	0.7842	0.2059	0
0.2059	0	0	0.7842	0.2059	0	0.7842	0
0.7842	0	0.2059	0	0.7842	0	0	0.2059
0	0.2059	0.7842	0	0	0.2059	0	0.7842
0	0	0.2059	0.7842	0	0.2059	0	0.7842
0.2059	0	0.7842	0	0.2059	0.7842	0	0
0.7842	0.2059	0	0	0.7842	0	0.2059	0
0	0.7842	0	0.2059	0	0	0.7842	0.2059
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.9. A-optimal blocked Design 3 when *a* = 0

B1				B2			
0	0.764942	0	0.235058	0	0.764942	0.235058	0
0.235058	0	0	0.764942	0.235058	0	0.764942	0
0.764942	0	0.235058	0	0.764942	0	0	0.235058
0	0.235058	0.764942	0	0	0.235058	0	0.764942
0	0	0.235058	0.764942	0	0.235058	0	0.764942
0.235058	0	0.764942	0	0.235058	0.764942	0	0
0.764942	0.235058	0	0	0.764942	0	0.235058	0
0	0.764942	0	0.235058	0	0	0.764942	0.235058
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.10. E-optimal blocked Design 3 when *a* = 0

B_1				B_2			
0	0.764942	0	0.346712	0	0.657281	0.346712	0
0.346712	0	0	0.657281	0.346712	0	0.657281	0
0.657281	0	0.346712	0	0.657281	0	0	0.346712
0	0.346712	0.657281	0	0	0.346712	0	0.657281
0	0	0.346712	0.657281	0	0.346712	0	0.657281
0.346712	0	0.657281	0	0.346712	0.657281	0	0
0.657281	0.346712	0	0	0.657281	0	0.346712	0
0	0.657281	0	0.346712	0	0	0.657281	0.346712
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal block designs obtained from Design 1 for the Reduced Cubic Canonical Model are as shown in Tables 3.11, 3.12 and 3.13, respectively.

Table 3.11. D-optimal blocked Design 1 when *c* = 0

B_1				B_2			
0.1307	0.7386	0	0.1307	0.1307	0.1307	0	0.7386
0.7386	0	0.1307	0.1307	0.7386	0.1307	0.1307	0
0	0.1307	0.1307	0.7386	0	0.7386	0.1307	0.1307
0.1307	0.1307	0.7386	0	0.1307	0	0.7386	0.1307
0.1307	0	0.1307	0.7386	0.1307	0	0.7386	0.1307
0.7386	0.1307	0.1307	0	0.7386	0.1307	0	0.1307
0	0.1307	0.7386	0.1307	0	0.1307	0.1307	0.7386
0.1307	0.7386	0	0.1307	0.1307	0.7386	0.1307	0
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.12. A-optimal blocked Design 1 when *c* = 0

B_1				B_2			
0.154846	0.690308	0	0.154846	0.154846	0.154846	0	0.690308
0.690308	0	0.154846	0.154846	0.690308	0.154846	0.154846	0
0	0.154846	0.154846	0.690308	0	0.690308	0.154846	0.154846
0.154846	0.154846	0.690308	0	0.154846	0	0.690308	0.154846
0.154846	0	0.154846	0.690308	0.154846	0	0.690308	0.154846
0.690308	0.154846	0.154846	0	0.690308	0.154846	0	0.154846
0	0.154846	0.690308	0.154846	0	0.154846	0.154846	0.690308
0.154846	0.690308	0	0.154846	0.154846	0.690308	0.154846	0
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.13. E-optimal blocked Design 1 when *c* = 0

B_1				B_2			
0.174178	0.651645	0	0.174178	0.174178	0.174178	0	0.651645
0.651645	0	0.174178	0.174178	0.651645	0.174178	0.174178	0
0	0.174178	0.174178	0.651645	0	0.651645	0.174178	0.174178
0.174178	0.174178	0.651645	0	0.174178	0	0.651645	0.174178
0.174178	0	0.174178	0.651645	0.174178	0	0.651645	0.174178
0.651645	0.174178	0.174178	0	0.651645	0.174178	0	0.174178
0	0.174178	0.651645	0.174178	0	0.174178	0.174178	0.651645
0.174178	0.651645	0	0.174178	0.174178	0.651645	0.174178	0
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal block designs obtained from Design 2 for Reduced Cubic Canonical Model are as shown in Tables 3.14, 3.15 and 3.16, respectively.

Table 3.14. D-optimal blocked Design 2 when *c* = 0

B_1				B_2			
0.1307	0.7386	0	0.1307	0.1307	0.1307	0	0.7386
0.7386	0	0.1307	0.1307	0.7386	0.1307	0.1307	0
0	0.1307	0.1307	0.7386	0	0.7386	0.1307	0.1307
0.1307	0.1307	0.7386	0	0.1307	0	0.7386	0.1307
0.1307	0.1307	0.7386	0	0.1307	0.7386	0.1307	0
0.7386	0.1307	0	0.1307	0.7386	0	0.1307	0.1307
0	0.7386	0.1307	0.1307	0	0.1307	0.7386	0.1307
0.1307	0	0.1307	0.7386	0.1307	0.1307	0	0.7386
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.15. A-optimal blocked Design 2 when *c* = 0

B_1				B_2			
0.154846	0.690308	0	0.154846	0.154846	0.154846	0	0.690308
0.690308	0	0.154846	0.154846	0.690308	0.154846	0.154846	0
0	0.154846	0.154846	0.690308	0	0.690308	0.154846	0.154846
0.154846	0.154846	0.690308	0	0.154846	0	0.690308	0.154846
0.154846	0.154846	0.690308	0	0.154846	0.690308	0.154846	0
0.690308	0.154846	0	0.154846	0.690308	0	0.154846	0.154846
0	0.690308	0.154846	0.154846	0	0.154846	0.690308	0.154846
0.154846	0	0.154846	0.690308	0.154846	0.154846	0	0.690308
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.16. E-optimal blocked Design 2 when c = 0

B1				B2			
0.174178	0.651645	0	0.174178	0.174178	0.174178	0	0.651645
0.651645	0	0.174178	0.174178	0.651645	0.174178	0.174178	0
0	0.174178	0.174178	0.651645	0	0.651645	0.174178	0.174178
0.174178	0.174178	0.651645	0	0.174178	0	0.651645	0.174178
0.174178	0.174178	0.651645	0	0.174178	0.651645	0.174178	0
0.651645	0.174178	0	0.174178	0.651645	0	0.174178	0.174178
0	0.651645	0.174178	0.174178	0	0.174178	0.651645	0.174178
0.174178	0	0.174178	0.651645	0.174178	0.174178	0	0.651645
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal block designs obtained from Design 3 for the Reduced Cubic Canonical Model are as shown in Tables 3.17, 3.18 and 3.19, respectively.

Table 3.17. D-optimal blocked Design 3 when c = 0

B1				B2			
0.1307	0	0.1307	0.7386	0.1307	0	0.7386	0.1307
0.7386	0.1307	0.1307	0	0.7386	0.1307	0	0.1307
0	0.1307	0.7386	0.1307	0	0.1307	0.1307	0.7386
0.1307	0.7386	0	0.1307	0.1307	0.7386	0.1307	0
0.1307	0.1307	0.7386	0	0.1307	0.7386	0.1307	0
0.7386	0.1307	0	0.1307	0.7386	0	0.1307	0.1307
0	0.7386	0.1307	0.1307	0	0.1307	0.7386	0.1307
0.1307	0	0.1307	0.7386	0.1307	0.1307	0	0.7386
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.18. A-optimal blocked Design 3 when c = 0

B1				B2			
0.154846	0	0.154846	0.690308	0.154846	0	0.690308	0.154846
0.690308	0.154846	0.154846	0	0.690308	0.154846	0	0.154846
0	0.154846	0.690308	0.154846	0	0.154846	0.154846	0.690308
0.154846	0.690308	0	0.154846	0.154846	0.690308	0.154846	0
0.154846	0.154846	0.690308	0	0.154846	0.690308	0.154846	0
0.690308	0.154846	0	0.154846	0.690308	0	0.154846	0.154846
0	0.690308	0.154846	0.154846	0	0.154846	0.690308	0.154846
0.154846	0	0.154846	0.690308	0.154846	0.154846	0	0.690308
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.19. E-optimal blocked Design 3 when c = 0

B_1				B_2			
0.174178	0	0.174178	0.651645	0.174178	0	0.651645	0.174178
0.651645	0.174178	0.174178	0	0.651645	0.174178	0	0.174178
0	0.174178	0.651645	0.174178	0	0.174178	0.174178	0.651645
0.174178	0.651645	0	0.174178	0.174178	0.651645	0.174178	0
0.174178	0.174178	0.651645	0	0.174178	0.651645	0.174178	0
0.651645	0.174178	0	0.174178	0.651645	0	0.174178	0.174178
0	0.651645	0.174178	0.174178	0	0.174178	0.651645	0.174178
0.174178	0	0.174178	0.651645	0.174178	0.174178	0	0.651645
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

3.5. Optimal Orthogonally Blocked Four Component Mixture Designs

When an equal number of observations are made, the two blocks in all three designs do not satisfy the orthogonality conditions in (3.5). But if we assume $a < b < c$, then the orthogonality conditions given in (3.5) are satisfied and we have orthogonal design in two blocks. Moreover, since the blocks in all three designs are orthogonal, we need not consider the process variables Z while optimizing the designs. These designs are useful in situations when the response of interest involves process variables.

Table 3.1. Portion of X Matrix along with the column sums arising from B_1 in Design 1

	1	2	3	4	12(1-2)	13(1-3)	14(1-4)	23(2-3)	24(2-4)	34(3-4)
	a	b	c	a	$ab(b\text{-}a)$	$ac(c\text{-}a)$	0	$bc(c\text{-}b)$	$ab(b\text{-}a)$	$ac(c\text{-}a)$
	b	c	a	a	$bc(c\text{-}b)$	$ab(b\text{-}a)$	$ab(b\text{-}a)$	$ac(c\text{-}a)$	$ac(c\text{-}a)$	0
	c	a	a	b	$ac(c\text{-}a)$	$ac(c\text{-}a)$	$bc(c\text{-}b)$	0	$ab(b\text{-}a)$	$ab(b\text{-}a)$
	a	a	b	c	0	$ab(b\text{-}a)$	$ac(c\text{-}a)$	$ab(b\text{-}a)$	$ac(c\text{-}a)$	$bc(c\text{-}b)$
	a	c	a	b	$ac(c\text{-}a)$	0	$ab(b\text{-}a)$	$ac(c\text{-}a)$	$bc(c\text{-}b)$	$ab(b\text{-}a)$
	b	a	a	c	$ab(b\text{-}a)$	$ab(b\text{-}a)$	$bc(c\text{-}b)$	0	$ac(c\text{-}a)$	$ac(c\text{-}a)$
	c	a	b	a	$ac(c\text{-}a)$	$bc(c\text{-}b)$	$ac(c\text{-}a)$	$ab(b\text{-}a)$	0	$ab(b\text{-}a)$
	a	b	c	a	$ab(b\text{-}a)$	$ac(c\text{-}a)$	0	$bc(c\text{-}b)$	$ab(b\text{-}a)$	$ac(c\text{-}a)$
	1/4	1/4	1/4	1/4	0	0	0	0	0	0
Column sum	A	A	A	A	B	B	C	C	B	B

where,

A = 4*a* + 2*b* + 2*c* + 1/4

B = 3*ab* (*b*-*a*) + 3*ac* (*c*-*a*) + *bc* (*c*-*b*)

C = 2*ab* (*b*-*a*) + 2*ac* (*c*-*a*) + 2*bc* (*c*-*b*)

For Design 1, the cross product column sums arising from B_1 are as given in Table 3.1. For B_2, column sums are the same as those obtained for B_1. Similarly, the column sums arising from two blocks for Design 2 and Design 3 are A, A, A, A, C, B, B, B, B, C and A, A, A, A, B, C, B, B, C, B, respectively.

For the Reduced Cubic Canonical Model, the form of matrix $\boldsymbol{X}$ for Design 1 with the condition $a < b < c$ is as given in (3.19).

$$\boldsymbol{X} = \begin{bmatrix}
a & b & c & a & ab(b-a) & ac(c-a) & 0 & bc(c-b) & ba(b-a) & ca(c-a) \\
b & c & a & a & bc(c-b) & ba(b-a) & ba(b-a) & ca(c-a) & ca(c-a) & 0 \\
c & a & a & b & ca(c-a) & ca(c-a) & cb(c-b) & 0 & ab(b-a) & ab(b-a) \\
a & a & b & c & 0 & ab(b-a) & ac(c-a) & ab(b-a) & ac(c-a) & bc(c-b) \\
a & c & a & b & ac(c-a) & 0 & ab(b-a) & ca(c-a) & cb(c-b) & ab(b-a) \\
b & a & a & c & ba(b-a) & ba(b-a) & bc(c-b) & 0 & ac(c-a) & ac(c-a) \\
c & a & b & a & ca(c-a) & cb(c-b) & ca(c-a) & ab(b-a) & 0 & ba(b-a) \\
a & b & c & a & ab(b-a) & ac(c-a) & 0 & bc(c-b) & ba(b-a) & ca(c-a) \\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0 \\
a & a & c & b & 0 & ac(c-a) & ab(b-a) & ac(c-a) & ab(b-a) & cb(c-b) \\
b & a & a & c & ba(b-a) & ba(b-a) & bc(c-b) & 0 & ac(c-a) & ac(c-a) \\
c & b & a & a & cb(c-b) & ca(c-a) & ca(c-a) & ba(b-a) & ba(b-a) & 0 \\
a & c & b & a & ac(c-a) & ab(b-a) & 0 & cb(c-b) & ca(c-a) & ba(b-a) \\
a & c & b & a & ac(c-a) & ab(b-a) & 0 & cb(c-b) & ca(c-a) & ba(b-a) \\
b & a & c & a & ba(b-a) & bc(c-b) & ba(b-a) & ac(c-a) & 0 & ca(c-a) \\
c & a & a & b & ca(c-a) & ca(c-a) & cb(c-b) & 0 & ab(b-a) & ab(b-a) \\
a & b & a & c & ab(b-a) & 0 & ac(c-a) & ba(b-a) & bc(c-b) & ac(c-a) \\
\frac{1}{4} & \frac{1}{4} & \frac{1}{4} & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix} \tag{3.19}$$

The forms of $|\boldsymbol{X}'\boldsymbol{X}|$ for Design 1, Design 2 and Design 3 are as given in (3.20), (3.21) and (3.22), respectively.

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccccc} P & R & R & Q & S & S & U & V & T & T \\ R & P & Q & R & S & T & V & U & S & T \\ R & Q & P & R & T & S & V & U & T & S \\ Q & R & R & P & T & T & U & V & S & S \\ \hline S & S & T & T & A & B & C & C & B & F \\ S & T & S & T & B & A & C & C & F & B \\ U & V & V & U & C & C & G & H & C & C \\ V & U & U & V & C & C & H & G & C & C \\ T & S & T & S & B & F & C & C & A & B \\ T & T & S & S & F & B & C & C & B & A \end{array}\right| \tag{3.20}$$

\

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccccc} P & Q & R & R & U & S & S & T & T & V \\ Q & P & R & R & U & T & T & S & S & V \\ R & R & P & Q & V & S & T & S & T & U \\ R & R & Q & P & V & T & S & T & S & U \\ \hline U & U & V & V & G & C & C & C & C & H \\ S & T & S & T & C & A & B & B & F & C \\ S & T & T & S & C & B & A & F & B & C \\ T & S & S & T & C & B & F & A & B & C \\ T & S & T & S & C & F & B & B & A & C \\ V & V & U & U & H & C & C & C & C & G \end{array}\right| \tag{3.21}$$

$$\boldsymbol{X'X}=\left|\begin{array}{cccc|cccccc} P & R & Q & R & S & U & S & T & V & T \\ R & P & R & Q & S & V & T & S & U & T \\ Q & R & P & R & T & U & T & S & V & S \\ R & Q & R & P & T & V & S & T & U & S \\ \hline S & S & T & T & A & C & B & B & C & F \\ U & V & U & V & C & G & C & C & H & C \\ S & T & T & S & B & C & A & F & C & B \\ T & S & S & T & B & C & F & A & C & B \\ V & U & V & U & C & H & C & C & G & C \\ T & T & S & S & F & C & B & B & C & A \end{array}\right| \tag{3.22}$$

where the expressions for P, Q, R, A and G are as the same as given in (3.10) and

$$S = -3ab^2(a-b) - 3a^2b(a-b) - 3ac^2(a-c) - 3a^2c(a-c) - bc^2(b-c) - b^2c(b-c)$$

$$U = -2ab^2(a-b) - 2a^2b(a-b) - 2ac^2(a-c) - 2a^2c(a-c) - 2bc^2(b-c) - 2b^2c(b-c)$$

$$V = -2a^2b(a-b) - 2a^2c(a-c) - 2abc(a-b) - 2abc(a-c) - 4abc(b-c)$$

$$\begin{aligned} B = {} & 2a^2b^2(a-b)^2 + 2a^2c^2(a-c)^2 - 4a^2(a-b)b(a-c)c + 2a(a-b)b^2(b-c)c \\ & + 2ab(a-c)(b-c)c^2 \end{aligned}$$

$$\begin{aligned} C = {} & a^2b^2(a-b)^2 + 2a^2(a-b)b(a-c)c + 3a(a-b)b^2(b-c)c + a^2(a-c)^2c^2 \\ & + 3ab(a-c)(b-c)c^2 \end{aligned}$$

$$F = 12a^2(a-b)b(a-c)c$$

$$H = 8a^2(a-b)b(a-c)c \tag{3.23}$$

$$\begin{aligned} |\boldsymbol{X'X}| = {} & 2304(a-b)^{10}(b-c)^6(ab+ac+bc-c^2)^4(2a^2-ab-3ac-bc+c^2)^6(2a^2b- \\ & 2ab^2+2a^2c+b^2c-2ac^2-bc^2)^2 \end{aligned} \tag{3.24}$$

$$T = T_1 / T_2 \tag{3.25}$$

where,

$$\begin{aligned} T_2 = {} & 12(a-b)^2(b-c)^2(ab+ac+bc-c^2)^2(-2a^2+ab+3ac+bc-c^2)^2(-2a^2b+ \\ & 2ab^2-2a^2c-b^2c+2ac^2+bc^2)^2 \end{aligned}$$

The expressions of T_1 and the eigenvalues λ_i (i = 1, 2, …, 10) are very lengthy and are available in the attached C.D. For optimal orthogonally blocked designs, we observe on using Mathematica with intervals of length 0.1, 0.01 and 0.05 that the D-, A- and E-optimal values of the three designs are obtained at the point $a = 0$. We have not considered the case $c = 0$ since the orthogonality conditions given in (3.5) are being satisfied when $a < b < c$. The forms of $\boldsymbol{X'X}$ for Design 1, Design 2 and Design 3 are as given in (3.20), (3.21) and (3.22), respectively with the following modifications.

$$P = \frac{1}{8} + 4b^2 + 4c^2$$

$$Q = \frac{1}{8} + 4bc$$

$$R = \frac{1}{8} + 2bc$$

$$S = -b^2(b-c)c - b(b-c)c^2$$

$$U = -2b^2(b-c)c - 2b(b-c)c^2$$

$$A = 2b^2(b-c)^2c^2$$

$$G = 4b^2(b-c)^2c^2$$

$$T = V = B = C = F = H = 0 \qquad (3.26)$$

For all the three designs, we have obtained the following results.

$$|\boldsymbol{X'X}| = 2304b^{10}(b-c)^6(bc-c^2)^4(-bc+c^2)^6(b^2c-bc^2)^2 \qquad (3.27)$$

$$T = T_1 / T_2 \qquad (3.28)$$

where,

$$T_1 = 37b^2 + 36b^4 - 46bc + 37c^2 - 72b^2c^2 + 14b^4c^2 + 24b^6c^2 - 28b^3c^3 - 96b^5c^3 + 36c^4 + 14b^2c^4 + 144b^4c^4 - 96b^3c^5 + 24b^2c^6$$

$$T_2 = 12b^2(b-c)^4c^2$$

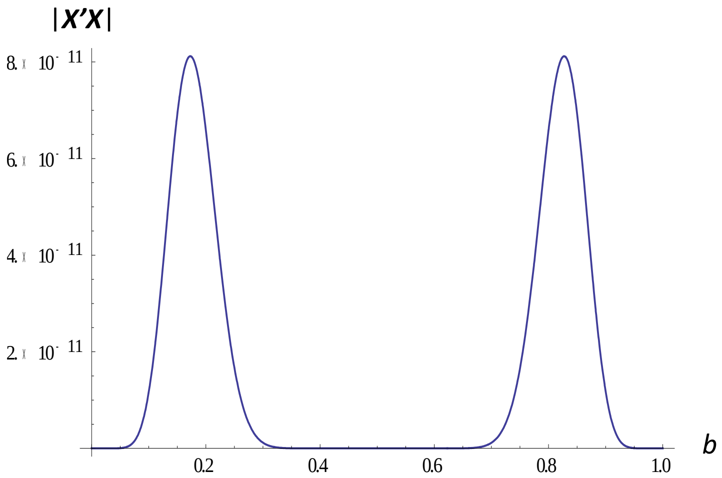


Figure 3.7. Graph of |*X′X*| against *b* for the Reduced Cubic Canonical Model

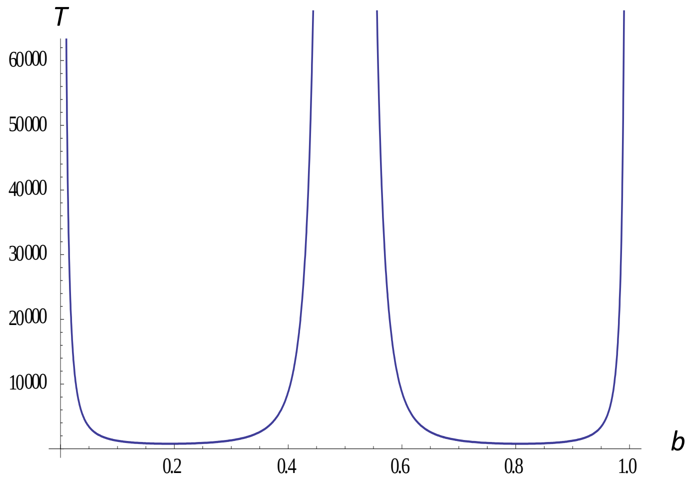


Figure 3.8. Graph of *T* against *b* for the Reduced Cubic Canonical Model

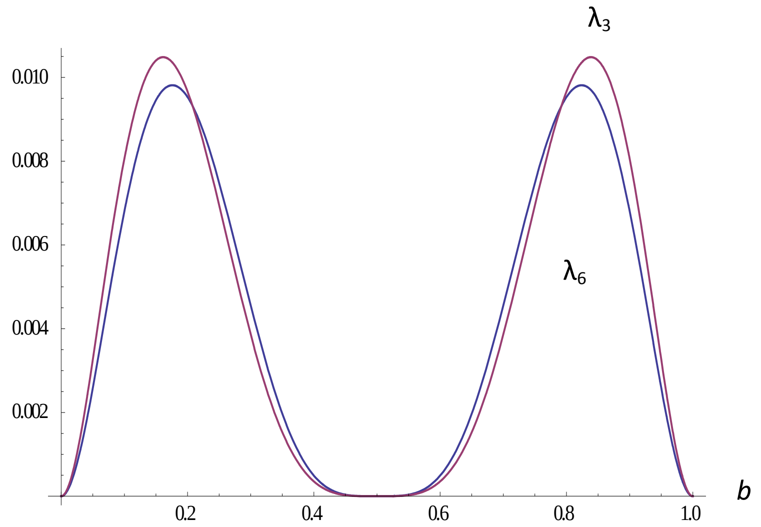


Figure 3.9. Graphs of eigenvalues λ_3 and λ_6 against *b* for the Reduced Cubic Canonical Model

The expressions of eigenvalues λ_i (i = 1, 2, …, 10) are very lengthy and are available in the attached C.D. Figures 3.7, 3.8 and 3.9, respectively represents the graphs of $|\boldsymbol{X'X}|$, T and eigenvalues λ_3 and λ_6 against b for the Reduced Cubic Canonical Model in the presence of orthogonal blocking.

We observe both numerically and graphically that

1. $|\boldsymbol{X'X}| = 0$ when $b = 0, \frac{1}{2}$ or 1.
2. The curve of $|\boldsymbol{X'X}|$ is *m*-shaped curve. Its maximum (= 8.11923×10^{-11}) is attained, only at the point $b = 0.172667$ ($a < b < c$).
3. T attains its minimum (= 775.656) when $b = 0.192039$ in.
4. $\lambda_0 = \min(\lambda_3, \lambda_6)$ attains its absolute maximum (= 0.00932) at $b = 0.208$.

The D, A and E-optimal orthogonal block designs obtained from Design 1 for the Reduced Cubic Canonical Model are as shown in Tables 3.20, 3.21 and 3.22, respectively.

Table 3.20. D-optimal orthogonally blocked Design 1

B_1				B_2			
0	0.1727	0.8274	0	0	0	0.8274	0.1727
0.1727	0.8274	0	0	0.1727	0	0	0.8274
0.8274	0	0	0.1727	0.8274	0.1727	0	0
0	0	0.1727	0.8274	0	0.8274	0.1727	0
0	0.8274	0	0.1727	0	0.8274	0.1727	0
0.1727	0	0	0.8274	0.1727	0	0.8274	0
0.8274	0	0.1727	0	0.8274	0	0	0.1727
0	0.1727	0.8274	0	0	0.1727	0	0.8274
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.21. A-optimal orthogonally blocked Design 1

B_1				B_2			
0	0.192039	0.807961	0	0	0	0.807961	0.192039
0.192039	0.807961	0	0	0.192039	0	0	0.807961
0.807961	0	0	0.192039	0.807961	0.192039	0	0
0	0	0.192039	0.807961	0	0.807961	0.192039	0
0	0.807961	0	0.192039	0	0.807961	0.192039	0
0.192039	0	0	0.807961	0.192039	0	0.807961	0
0.807961	0	0.192039	0	0.807961	0	0	0.192039
0	0.192039	0.807961	0	0	0.192039	0	0.807961
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.22. E-optimal orthogonally blocked Design 1

B_1				B_2			
0	0.208	0.792	0	0	0	0.792	0.208
0.208	0.792	0	0	0.208	0	0	0.792
0.792	0	0	0.208	0.792	0.208	0	0
0	0	0.208	0.792	0	0.792	0.208	0
0	0.792	0	0.208	0	0.792	0.208	0
0.208	0	0	0.792	0.208	0	0.792	0
0.792	0	0.208	0	0.792	0	0	0.208
0	0.208	0.792	0	0	0.208	0	0.792
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal orthogonal block designs obtained from Design 2 for the Reduced Cubic Canonical Model are as shown in Tables 3.23, 3.24 and 3.25, respectively.

Table 3.23. D-optimal orthogonally blocked Design 2

B_1				B_2			
0	0.1727	0.8274	0	0	0	0.8274	0.1727
0.1727	0.8274	0	0	0.1727	0	0	0.8274
0.8274	0	0	0.1727	0.8274	0.1727	0	0
0	0	0.1727	0.8274	0	0.8274	0.1727	0
0	0	0.1727	0.8274	0	0.1727	0	0.8274
0.1727	0	0.8274	0	0.1727	0.8274	0	0
0.8274	0.1727	0	0	0.8274	0	0.1727	0
0	0.8274	0	0.1727	0	0	0.8274	0.1727
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.24. A-optimal orthogonally blocked Design 2

B_1				B_2			
0	0.192039	0.807961	0	0	0	0.807961	0.192039
0.192039	0.807961	0	0	0.192039	0	0	0.807961
0.807961	0	0	0.192039	0.807961	0.172665	0	0
0	0	0.192039	0.807961	0	0.807961	0.192039	0
0	0	0.192039	0.807961	0	0.192039	0	0.807961
0.192039	0	0.807961	0	0.192039	0.807961	0	0
0.807961	0.192039	0	0	0.807961	0	0.192039	0
0	0.807961	0	0.192039	0	0	0.807961	0.192039
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.25. E-optimal orthogonally blocked Design 2

B_1				B_2			
0	0.208	0.792	0	0	0	0.792	0.208
0.208	0.792	0	0	0.208	0	0	0.792
0.792	0	0	0.208	0.792	0.208	0	0
0	0	0.208	0.792	0	0.792	0.208	0
0	0	0.208	0.792	0	0.208	0	0.792
0.208	0	0.792	0	0.208	0.792	0	0
0.792	0.208	0	0	0.792	0	0.208	0
0	0.792	0	0.208	0	0	0.792	0.208
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

The D, A and E-optimal orthogonal block designs obtained from Design 3 for the Reduced Cubic Canonical Model are as shown in Tables 3.26, 3.27 and 3.28, respectively.

Table 3.26. D-optimal orthogonally blocked Design 3

B_1				B_2			
0	0.8274	0	0.1727	0	0.8274	0.1727	0
0.1727	0	0	0.8274	0.1727	0	0.8274	0
0.8274	0	0.1727	0	0.8274	0	0	0.1727
0	0.1727	0.8274	0	0	0.1727	0	0.8274
0	0	0.1727	0.8274	0	0.1727	0	0.8274
0.1727	0	0.8274	0	0.1727	0.8274	0	0
0.8274	0.1727	0	0	0.8274	0	0.1727	0
0	0.8274	0	0.1727	0	0	0.8274	0.1727
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.27. A-optimal orthogonally blocked Design 3

B_1				B_2			
0	0.827371	0	0.192039	0	0.807961	0.192039	0
0.192039	0	0	0.827371	0.192039	0	0.807961	0
0.827371	0	0.192039	0	0.807961	0	0	0.192039
0	0.192039	0.827371	0	0	0.192039	0	0.807961
0	0	0.192039	0.827371	0	0.192039	0	0.807961
0.192039	0	0.827371	0	0.192039	0.807961	0	0
0.827371	0.192039	0	0	0.807961	0	0.192039	0
0	0.827371	0	0.192039	0	0	0.807961	0.192039
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.28. E-optimal orthogonally blocked Design 3

B_1				B_2			
0	0.792	0	0.208	0	0.792	0.208	0
0.208	0	0	0.792	0.208	0	0.792	0
0.792	0	0.208	0	0.792	0	0	0.208
0	0.208	0.792	0	0	0.208	0	0.792
0	0	0.208	0.792	0	0.208	0	0.792
0.208	0	0.792	0	0.208	0.792	0	0
0.792	0.208	0	0	0.792	0	0.208	0
0	0.792	0	0.208	0	0	0.792	0.208
1/4	1/4	1/4	1/4	1/4	1/4	1/4	1/4

Table 3.29. The numerical values of the design parameters for four component mixtures based on F-squares for the Reduced Cubic Canonical Model

Optimality Criteria	Optimal blocked		Optimal orthogonally blocked
	a = 0, c = 1 - b	c = 0, a = 0.5 – b/2	a = 0, c = 1 - b
	b	b	b
D - optimality	0.205902, 0.784231	0.738560	0.172665, 0.827371
A - optimality	0.235058, 0.764942	0.690308	0.192039, 0.807961
E - optimality	0.346712, 0.657281	0.651645	0.208000, 0.792000

Table 3.29 depicts the values of parameters a, b and c for the Reduced Cubic Canonical model in four components for optimal blocked and optimal orthogonally blocked designs presented in Sections 3.4 and 3.5, respectively.

3.6. Conclusions

In this chapter, we have constructed optimal designs as well as optimal orthogonally blocked designs in two blocks based on F-squares for the Reduced Cubic Canonical Model (3.2) in four components. From the results in Section 3.4, we infer that when no process variables are involved, Design 1, Design 2 and Design 3 are D-, A- and E-optimal for the case a = 0 at the particular values of b at 0.205902, 0.235058 and 0.346712, respectively. The same for the case c = 0 are obtained at the values of b being 0.73856, 0.690308 and 0.651645, respectively.

From the results in Section 3.5, the D-, A- and E-optimal orthogonally blocked designs in the presence of process variables are obtained at b = 0.172665, 0.192039 and 0.208, respectively for a = 0 and c = 0.827371, 0.807961 and 0.792, respectively.

Chapter 4

UNIFORM DESIGNS BASED ON F-SQUARES AND MEASURES OF DIFFERENT DISCREPANCIES

4.1. Introduction

Fang (1980) obtained uniform designs (UD) by applying number theoretic methods to experimental designs. Since then there has been a tremendous research on uniform designs which are space filling designs and are robust to the underlying model assumptions. One of the several benefits of performing UD is that it helps to study the relationships between the factors and the response of interest using economical number of runs. For s factors and n runs without loss of generality, the experimental domain is assumed to be the unit cube $C^s = [0, 1]^s$. Uniform designs (UDs) scatter the n experimental points uniformly over C^s. Discrepancy measure given by Warnock (1972) is adopted to choose the n points with the smallest discrepancy.

Fang et al. (1999) obtained uniform designs based on latin squares. Some practical considerations require the presence of two or more experimental components to be present in equal values. Gold forms alloys with most metals but for jewelry, the most common alloying metals are Silver, Copper and Zinc. White Gold (18 k) contains Gold 75%, Palladium 10%, Nickel 10% and Zinc 5%. Here, Palladium and Nickel are present in equal proportions. Consider the case of Nickel-Copper alloy viz., Ni-Cu 400 (Max.) which is resistant to seawater and steam at high temperatures as well as salt and caustic solutions. This particular alloy is characterized by good corrosion resistance, good weld ability and moderate to high strength and hence finds applications in the chemical, marine and oil industries. The chemical requirements (% by weight) of Ni-Cu 400 (Max.) are as follows:

C	0.30
Mn	1.00
Si	0.30
Ni	65.00
Cu	31.50
Fe	1.90

This facilitates the use of F-squares rather than the usual Latin square. Aggarwal et al. (2009) obtained mixture designs in orthogonal blocks using F-squares.

In this chapter, we have obtained uniform designs based on cyclic F-squares. Section 4.2 presents the prerequisites on uniform designs as well as F-squares. Section 4.3

presents some measures of uniformity. For the two difference schemes, viz, DS_1 and DS_2 defined in Section 4.4, we study the properties of UF-type designs that may be used to reduce the computing time. Section 4.5 gives the TA algorithm followed by some numerical comparisons between G-Uniform designs and F-square designs followed by comparisons between latin square based designs and F-uniform designs. Wrap- around discrepancy is calculated in Section 4.6. Conclusions and additional discussions are dealt with in the last section. We have proposed a subset of UL-type designs called the UF-type designs based on F-squares.

4.2. The Prerequisites on Uniform Designs as well as F-squares

Definition 4.1 A U-type design of size $n \times s$, denoted by $U_{n,s} = (u_{ij})$, is an $n \times s$ ($s \leq n$) matrix with rank s such that each column is a permutation of $\{1, 2, \ldots, n\}$. A U-type design $U_{n,s}$ provides an experimental design where there are s factors each having n levels within n experiments, (Fang and Wang (1981)). The induced matrix of $U_{n,s}$ is $X_{n,s} = (x_{ij})$, where

$$x_{ij} = \frac{u_{ij} - 0.5}{n} \tag{4.1}$$

for $i = 1, 2, \ldots, n$ and $j = 1, 2, \ldots, s$. The design points in $U_{n,s}$ and $X_{n,s}$ may be put in a one-to-one correspondence implying that they may be used interchangeably. The matrix $X_{n,s}$ may be considered as n points in C^s. A U-uniform design is $U_{n,s}$ with induced matrix $X_{n,s}$ having the smallest discrepancy. Equivalent U-type designs are the ones which may be obtained from each other by permuting rows and/or columns. Without loss of generality, the first column of a U-uniform design may always be taken as $(1, 2, \ldots, n)'$. There are (n! - 1) possible permutations for the second column, (n! - 2) choices for the third column and so on. The search for the best s columns might involve intolerable computing time even for moderate n and s. Fang and Wang (1981) presented the use of good lattice point (*glp*) sets in order to reduce the computing cost and time.

A UF-type design may be obtained by selecting s linearly independent columns of a cyclic F-square. Laywine (1989) obtained F-squares by making substitutions based on numbers for latin squares. For example, let us consider the following latin square of side 4.

Latin Square of order 4				**FSI(4) Square number 1**				**FSI(4) Square number 2**				**FSI(4) Square number 3**			
1	2	3	4	1	2	3	3	1	3	3	2	1	3	2	3
2	1	4	3	2	1	3	3	2	3	3	1	2	3	1	3
3	4	1	2	3	3	1	2	3	1	2	3	3	2	3	1
4	3	2	1	3	3	2	1	3	2	1	3	3	1	3	2

By substituting the number 4 = 3 in the latin square, FSI(4) is obtained. FSI(4) generates two distinct F-squares via permutations of the last three columns. F-squares are identified by simply writing down the first row of the square. Hence we will represent square number 2 obtained from FSI(4) by simply writing its first row as (1 3 3 2). Aggarwal et al. (2009) gave the following definitions.

Definition 4.2 An F-square with the first row and first column in natural order is called a standard F-square where by natural order we mean to imply that each element is followed by the same element (if it assumes an equal proportion) or the next element cyclically.

For four components, we have taken 4 = 3 as it yields minimum L_2-discrepancy, the natural order being 1 2 3 3. Hence for general q, the natural order is 1 2 … (q-1) (q-1) throughout for our calculations.

Definition 4.3 Two F-squares are equivalent if one can be derived from the other by permutations of rows and/or permutations of columns and/or permutations of elements.

Fang et al. (1999) obtained a subset of U-type designs based on cyclic Latin squares called the UL-type designs. In this paper, we have constructed another subset of U-type designs based on cyclic F-squares. This new subset of U- type designs called UF-type designs may be obtained by selecting linearly independent columns of cyclic F-squares. We use the threshold accepting (TA) algorithm proposed by Dueck and Scheuer (1990) and applied by Fang et al. (1999) in order to determine the “best” UF-type design. We denote the F-uniform design so obtained by $UF_n(n^s)$.

4.3. Uniform Designs

If there are s factors of interest on a standard domain C^s, then the purpose is to choose a set of n experimental points $\boldsymbol{X} = (x_1, x_2, \ldots, x_n) \subset C^s$ that is uniformly scattered on C^s. Let $M(\boldsymbol{X})$ be a measure of uniformity of $\boldsymbol{X}$ such that the smaller M corresponds to better uniformity. The following subsection presents some of the standard measures of uniformity available in literature.

4.3.1 Measures of Uniformity

Let $F(x)$ be the uniform distribution on C^s and $F_n(\boldsymbol{x})$ be the empirical distribution function of $\boldsymbol{X}$, i.e.,

$$F_n(x) = \frac{1}{n}\sum_{i=1}^{n} I\{x_i \le x\}, \tag{4.2}$$

where $I\{\cdot\}$ is the indicator function and all inequalities are understood to be component wise. The L_p-discrepancy of $\boldsymbol{X}$ is defined as

$$D_p(\mathcal{P}) = \left[\int_{C^s} \left|F_n(x) - F(x)\right|^p dx\right]^{\frac{1}{p}}, \tag{4.3}$$

where $F(x)$ is the distribution function of the uniform distribution over C^s. When $p = \infty$, $D \equiv D_\infty$ is also called the discrepancy (or star-discrepancy). This is perhaps the most universally used measure of discrepancy expressed as

$$D(\mathcal{P}) = sup_{x \in C^s}|F_n(x) - F(x)|. \tag{4.4}$$

No general algorithm is available for calculating discrepancy in multidimensional situations. Bundschuh and Zhu (1993) presented a method for exact calculation of the discrepancy of low dimensional finite point sets. Warnock (1972) gave the following analytic formula for calculating L_2-discrepancy

$$\left(D_2(\mathcal{P})\right)^2 = 3^{-s} - \frac{2^{1-s}}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}(1 - x_{kl}^2) + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left[1 - max\left(x_{ki}, x_{ji}\right)\right] \tag{4.5}$$

where $x_k = (x_{k1}, \ldots, x_{ks})$

Fang and Wang (1981) presented a note on uniform distribution and experimental design. They gave the following formula to compute the discrepancy of the G-Uniform design.

$$D(n,a) = \frac{1}{n}\sum_{k=1}^{n}\prod_{v=1}^{s}\left(1-\frac{2}{\pi}ln\left(2sin\pi\frac{a_{vk}}{n+1}\right)\right). \tag{4.6}$$

where $a_{vk} = a_v k(mod\ n)$, $1 \le a_{vk} < n$ for $1 \le k < n$ and $a_{vn} = n$.

Hickernell (1998a) gave three modified L_2-discrepancies, viz.; the symmetric L_2-discrepancy (SD_2), the centered L_2-discrepancy (CD_2) and modified L_2-discrepancy (MD_2). These uniformity measures are described in Fang et al. (2000). Hickernell (1998a) gave an analytical expression for the centered L_2-discrepancy as

$$\begin{aligned}\left(CD_2(\mathcal{P})\right)^2 = \left(\frac{13}{12}\right)^s - \frac{2}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}\left(1+\frac{1}{2}|x_{kl}-0.5|-\frac{1}{2}|x_{kl}-0.5|^2\right)\\ +\frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(1+\frac{1}{2}|x_{ki}-0.5|+\frac{1}{2}|x_{ji}-0.5|-\frac{1}{2}|x_{ki}-x_{ji}|\right).\end{aligned} \tag{4.7}$$

where $x_k = (x_{k1}, \ldots, x_{ks}) \in \mathcal{P}$, the centered L_2-discrepancy takes into account not only the uniformity of $\mathcal{P}$ over C^s, but also uniformity of all the projections of $\mathcal{P}$ over C^s.

4.4. U-type Designs Based on Cyclic F-Squares

Let $x_1, \ldots, x_n$ be the entries of an F-square. Define the left shift operator L on the F-square by $LF(x_1, \ldots, x_n) = (x_2, x_3, \ldots, x_n, x_1)$. We now present the following definitions.

Definition 4.4 A left cyclic F-Square (LCFS) of order n is an F-square of order n such that $x_{i+1} = LFx_i$, $i = 1, \ldots, n$-1, where x_i is the i^{th} row of the F-square.
Let $\mathcal{P}_n = \{(x_1, \ldots, x_n) : (x_1, \ldots, x_n)$ is a permutation of $\{1, \ldots, n\}\}$. An LCFS is uniquely determined by its first row. The LCFS with the first row $\alpha \in \mathcal{P}_n$ is denoted by $LCF(\alpha)$. Any s columns of an LCFS form a U-type design called a UF-type design.

Definition 4.5 A left cyclic standard F-Square (LCSFS) of order n is a standard F-square of order n such that $x_{i+1} = LFx_i$, $i = 1, \ldots, n$-1, where x_i is the i^{th} row of the standard F-square.

Let $\mathcal{P}_n = \{(x_1, \ldots, x_n) : (x_1, \ldots, x_n)$ is a permutation of $\{1, \ldots, n\}\}$. An LCSFS is uniquely determined by its first row. The LCSFS with the first row $\alpha \in \mathcal{P}_n$ is denoted by $LCF(\alpha)$. Any s columns of an LCSFS form a U-type design called a USF-type design. We now present the definition of difference sequence in the setup of F-squares.

Definition 4.6 Let $i_1, \ldots, i_s$ be s integers in the first row of the F-square such that $1 \leq i_1 \leq \ldots i_s \leq (n - 1) + i_1 + 1$. The sequence $\{i_2 - i_1, i_3 - i_2, \ldots, i_{s-1} - i_{s-2}, \ i_1 - i_s\}(mod\ (n-1))$ is called a difference sequence. If the sequence takes values in $\{0, 1\}$ then we denote it as DS_1. The standard F-squares shown in Table 4.4 conform to the sequence DS_1. We have obtained the "best" F-squares having the least L_2-discrepancy as shown in Table 4.5. If the difference sequence assumes values in $\{1, 2, \ldots, (n - 1)\}$, then we denote this sequence as DS_2. Note that the first row of the LCFS in Table 4.5 is not in standard form. Moreover if $\{d_1, \ldots, d_s\}$ is a difference sequence, then $\sum_{i=1}^{s} d_i = (n-1)$. For $\mathbf{K} = LCF(\alpha)$, let $\mathbf{K}(i_1, \ldots, i_s)$ be the submatrix consisting of the $i_1^{th}, \ldots, i_s^{th}$ columns of $\mathbf{K}$. Theorems 4.1 and 4.2 are applicable to both the difference sequences DS_1 and DS_2.

Theorem 4.1 If the indices of two submatrices $\mathbf{K}(i_1, \ldots, i_s)$ and $\mathbf{K}(j_1, \ldots, j_s)$ of $\mathbf{K} = CF(\boldsymbol{\alpha})$, $\alpha \in \mathcal{P}$, have the same difference sequence, then these two UF-type designs are equivalent.

Proof: Let $\{d_1, \ldots, d_s\}$ be the difference sequence of both the indices. We have to show $\boldsymbol{B} = LF(1, 1 + d_1, 1 + d_1 + d_2, \ldots, 1 + d_1 + \ldots + d_{s-1})$ and $\boldsymbol{A} = LF(i_1, \ldots, i_s)$ are equivalent. Trivially, $(i_1 + k)^{th}$ row of the $\boldsymbol{B}$ is just the $(i + k)^{th}$ row of $\boldsymbol{A}$.

Theorem 4.2 If $\boldsymbol{A}$ and $\boldsymbol{B}$ are two $n \times s$ submatrices of $\mathbf{K} = \mathrm{LCF}(\boldsymbol{\alpha})$, $\alpha \in \mathcal{P}$, with difference sequences $\{d_1, d_2, \ldots, d_s\}$ and $\{d_2, d_3, \ldots, d_s, d_1\}$, respectively, then $\mathbf{A}$ and $\mathbf{B}$ are equivalent.

Proof: From Theorem 4.1, we can assume
$\boldsymbol{A} = LF(1, 1 + d_1, 1 + d_1 + d_2, \ldots, 1 + d_1 + \ldots + d_{s-1})$ and

$\boldsymbol{B} = LF(1 + d_1, 1 + d_1 + d_2, \ldots, 1 + d_1 + \ldots + d_{s-1}, 1 + d_1 + \ldots + d_s)$

$\quad = LF(1 + d_1, 1 + d_1 + d_2, \ldots, 1 + d_1 + \ldots + d_{s-1}, 1 + (n - 1))$

$= LF(1 + d_1, 1 + d_1 + d_2, \ldots, 1 + d_1 + \ldots + d_{s-1}, 1)$

So that matrix ***A*** and ***B*** are equivalent.

FSII(4) generates six distinct F-squares (including itself) via permutations of the last three columns as shown in the Table 4.1. There is no change in L_2-discrepancy (11.717E-02) in all these F-squares.

Table 4.1. Squares obtained from FSII(4) by permuting the last three columns.

Square number 1				Square number 2				Square number 3			
1	2	3	3	1	2	3	3	1	3	2	3
2	3	3	1	2	3	1	3	2	3	3	1
3	3	1	2	3	3	2	1	3	1	3	2
3	1	2	3	3	1	3	2	3	2	1	3
Square number 4				**Square number 5**				**Square number 6**			
1	3	2	3	1	3	3	2	1	3	3	2
2	1	3	3	2	3	1	3	2	1	3	3
3	2	3	1	3	1	2	3	3	2	1	3
3	3	1	2	3	2	3	1	3	3	2	1

Table 4.2. LCSFS with difference sequence DS_1 for *n*=5

Square No.	First row of square	Square No.	First row of square	Square No.	First row of square
1	1 2 3 4 4	9	1 3 2 4 4	17	1 4 4 3 2
2	1 2 4 3 4	10	1 3 4 4 2	18	1 4 3 2 4
3	1 4 2 3 4	11	1 3 4 2 4	19	1 4 4 2 3
4	1 2 4 3 4	12	1 3 4 4 2	20	1 4 4 3 2
5	1 2 3 4 4	13	1 4 4 2 3	21	1 4 2 4 3
6	1 2 4 4 3	14	1 4 2 4 3	22	1 4 2 3 4
7	1 3 2 4 4	15	1 2 4 4 3	23	1 4 3 4 2
8	1 3 4 2 4	16	1 4 3 4 2	24	1 4 3 2 4

Table 4.3. LCFS with difference sequence DS_2 for *n*=5

Square No.	First row of square	Square No.	First row of square	Square No.	First row of square
1	1 4 3 4 2	9	1 2 3 4 4	17	1 2 4 3 4
2	1 4 2 4 3	10	1 3 2 4 4	18	1 2 4 4 3
3	1 4 3 4 2	11	1 2 3 4 4	19	1 3 4 2 4
4	1 4 2 4 3	12	1 3 2 4 4	20	1 4 2 3 4
5	1 4 4 3 2	13	1 3 4 4 2	21	1 3 4 2 4
6	1 4 4 2 3	14	1 2 4 4 3	22	1 4 2 3 4
7	1 4 4 3 2	15	1 2 4 3 4	23	1 4 3 2 4
8	1 4 4 2 3	16	1 2 4 4 3	24	1 4 3 2 4

The first row of the 24 distinct LCSFS with their common L_2-discrepancy are given in Table 4.2 for the difference sequence DS_1. Table 4.3 presents the LCFS for the difference sequence DS_2. L_2-discrepancy in Table 4.2 and 4.3 are 6.4817E-02 and 6.1394E-02, respectively.

4.5. F-Uniform Designs, Threshold Accepting Algorithm and Numerical Comparisons

To construct the "best" UF-type design, we use the two stage procedure presented by Fang et al. (1999) for the case of latin square based uniform designs.
First, an $\alpha^* \epsilon \mathcal{P}_n$ is chosen such that

$$D_2\big(LCF(\alpha^*)\big) = \min_{\alpha\in\mathcal{P}n} D_2\big(LCF(\alpha)\big) \tag{4.8}$$

Then for $K = LCF(\alpha^*)$, we find $(i_1^*, \dots, i_s^*)$ such as

$$D\big(K(i_1^*, \dots, i_s^*)\big) = \min_{1\le i_1<\dots<i_s\le n} D\big(K(i_1, \dots, i_s)\big).$$

The design $K(i_1^*, \dots, i_s^*)$ is called an F-uniform design. It is comparatively easier to calculate L_2-discrepancy (4.5). We use it in the first stage towards obtaining the 'best' UF-type design.

Dueck and Scheuer (1990) presented the TA algorithm to facilitate the search for the best α. Let χ be a set of finite elements and *f*(*x*) be an objective function that maps χ into the real numbers. We want to choose $x^* \epsilon\, \chi$ such that

$$f(x^*) = \min_{x \in \chi} f(x). \tag{4.9}$$

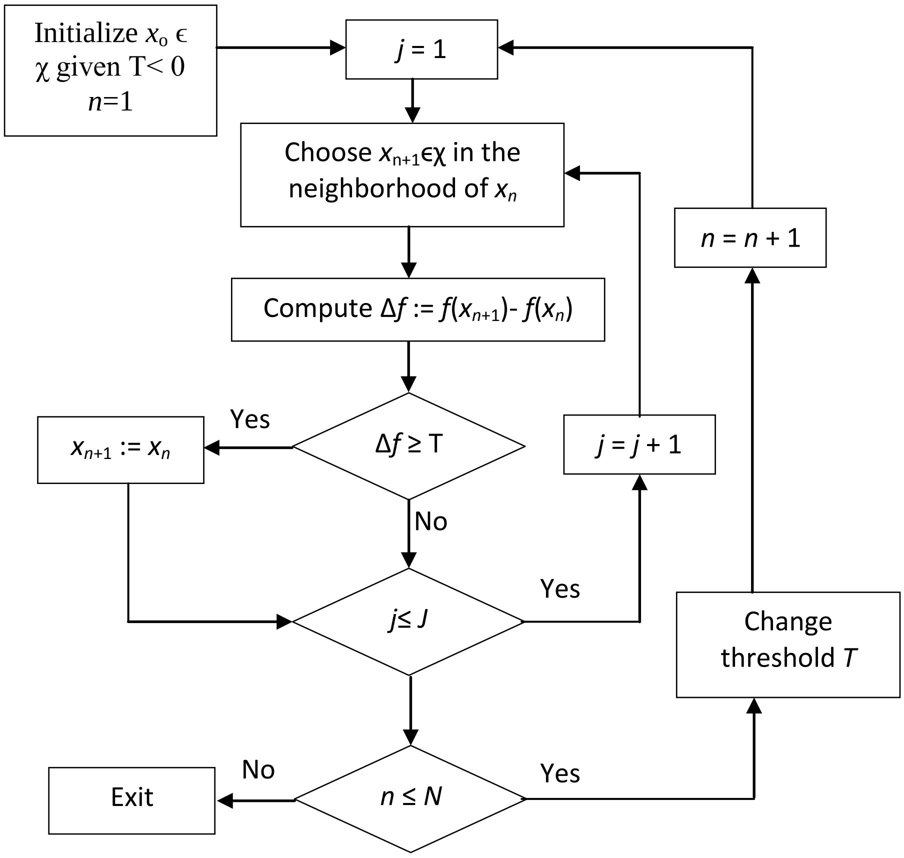


Figure 4.1. Flow chart of TA Algorithm

Suppose that for any $x \epsilon \chi$. Define a neighborhood $N(x)$ with respect to the domain χ and the objective function f. The TA algorithm begins with an arbitrary element χ in order to minimize a certain function over a finite set.

A local search algorithm is a natural way for solving problem (4.9). This algorithm starts with an initial element $x_0 \epsilon \chi$ which may be chosen randomly. In the n^{th} iteration, we replace the current solution x_n by the new one x_{n+1}. Figure 4.1 gives the flow chart of TA algorithm (Fang et al. (1999)).

In our case, χ is $\mathcal{P}_n$. By hit and trial method, we keep on changing the thresholds throughout our search towards attaining a local minimum. This search ultimately leads to the global minimum. The number of possible $\boldsymbol{\alpha}$ that need to be considered is reduced to $(n - 1)!$, since $LCF(\boldsymbol{\alpha})$ and $LCF(L^m\boldsymbol{\alpha})$ are obviously equivalent for m = 1,

…, n - 1. Let $\alpha = (a_1, \ldots, a_n) \in \chi$. A part of the final results are given in Table 4.4 and Table 4.5 for both the difference sequences DS_1 and DS_2.

If the rows of $\mathcal{P}_n$ are i.i.d. random vectors from the uniform distribution over C^s, then it is known that

$$E([D_2(\mathcal{P})]^2) = \frac{1}{n}\left(\frac{1}{2^s} - \frac{1}{3^s}\right). \tag{4.10}$$

Obviously $D_2(LCF(\boldsymbol{\alpha}))$ is much lower than the corresponding expected value of D_2 in (4.10). We have arrived at our results by programming the calculations on C++. The programs are available in the Appendix.

In the second stage, towards constructing the "best" UF-type designs, we have applied Theorems 4.1 and 4.2 for $n \leq 50$ and $s \leq 7$. We have presented our results till n = 32.

Figure 4.2 and Figure 4.5 present the decreasing plot of log (D_2(LCF(α))) against n for DS_1 and DS_2, respectively. Figure 4.3 and Figure 4.4 present the L_2-discrepancy against n for the difference sequences DS_1 and DS_2, respectively. Figure 4.6 presents $E([D_2(\mathcal{P})]^2)$ against n for s = 3 and 4 for DS_1.

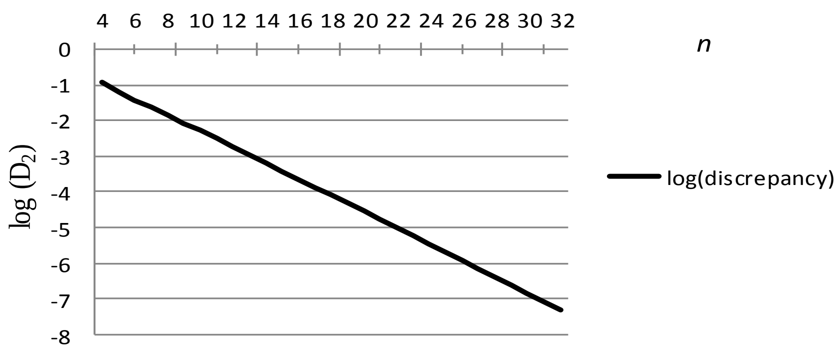


Figure-4.2. Plot of log (D_2) against *n* for DS_1.

Table 4.4. The first row of LCSFS with difference sequence DS_1

n	First Row of LCSFS	L_2-discrepancy
4	1 2 3 3	11.717E-02
5	1 2 3 4 4	6.481E-02
6	1 2 3 4 5 5	3.815E-02
7	1 2 3 4 5 6 6	2.311E-02
8	1 2 3 4 5 6 7 7	1.409E-02
9	1 2 3 4 5 6 7 8 8	8.577E-03
10	1 2 3 4 5 6 7 8 9 9	5.188E-03
11	1 2 3 4 5 6 7 8 9 10 10	3.118E-03
12	1 2 3 4 5 6 7 8 9 10 11 11	1.864E-03
13	1 2 3 4 5 6 7 8 9 10 11 12 12	1.109E-03
14	1 2 3 4 5 6 7 8 9 10 11 12 13 13	6.580E-04
15	1 2 3 4 5 6 7 8 9 10 11 12 13 14 14	3.892E-04
16	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 15	2.300E-04
17	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 16	1.350E-04
18	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 17	7.965E-05
19	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 18	4.686E-05
20	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 19	2.756E-05
21	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 20	1.621E-05
22	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 21	9.538E-06
23	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 22	5.613E-06
24	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 23	3.304E-06
25	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 24	1.946E-06
26	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 25	1.147E-06
27	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 26	6.765E-07
28	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 27	3.992E-07
29	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 28	2.357E-07
30	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 29	1.393E-07
31	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 30	8.241E-08
32	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 31	4.877E-08

Table 4.5. The first row of LCFS with difference sequence DS_2.

n	First Row of LCFS	L_2-discrepancy
4	1 2 3 3	11.717E-02
5	1 4 3 4 2	6.139E-02
6	1 3 5 5 2 4	3.510E-02
7	1 3 2 6 4 5 6	2.088E-02
8	1 5 2 7 7 4 3 6	1.276E-02
9	1 8 8 4 2 5 6 3 7	7.748E-03
10	1 7 3 9 4 6 9 8 5 2	4.717E-03
11	1 4 6 8 9 10 5 3 10 2 7	2.854E-03
12	1 7 10 4 5 11 9 11 2 3 6 8	1.717E-03
13	1 12 4 6 5 7 9 10 3 2 8 12 11	1.028E-03
14	1 8 10 4 11 12 9 13 3 2 6 13 5 7	6.130E-04
15	1 7 12 6 9 10 13 8 14 5 3 2 4 14 11	3.640E-04
16	1 9 8 2 15 6 7 12 15 11 14 10 5 3 4 13	2.160E-04
17	1 10 2 5 14 6 7 9 12 11 3 4 16 8 13 16 15	1.280E-04
18	1 7 17 17 13 5 14 12 3 16 6 8 9 11 4 10 2 15	7.538E-05
19	1 18 18 13 9 6 11 5 16 15 3 14 4 12 10 8 2 17 7	4.447E-05
20	1 16 15 18 19 5 6 8 2 13 4 10 17 11 3 12 14 7 9 19	2.622E-05
21	1 20 19 12 11 17 13 6 10 8 2 3 5 14 15 20 4 9 16 7 18	1.545E-05
22	1 16 14 8 17 15 7 10 21 2 4 5 13 19 6 9 3 12 11 18 20 21	9.105E-06
23	1 5 14 6 19 11 17 7 8 12 2 21 9 10 4 16 20 18 3 22 22 13 15	5.366E-06
24	1 16 23 5 21 13 6 9 14 22 2 23 17 12 20 19 15 10 7 11 18 4 8 3	3.163E-06
25	1 20 3 24 12 22 23 2 9 5 11 8 15 13 4 24 17 14 7 6 21 10 19 18 16	1.865E-06
26	1 3 21 13 17 25 8 6 19 23 15 14 9 7 22 18 12 25 20 16 5 11 4 10 2 24	1.101E-06
27	1 9 8 4 3 18 26 10 20 23 25 22 14 19 2 11 26 21 13 6 16 12 7 15 24 5 17	6.497E-07
28	1 25 6 16 13 17 20 26 18 11 21 27 23 19 4 10 24 3 15 9 5 8 14 12 7 22 27 2	3.837E-07
29	1 12 27 5 20 4 17 2 15 6 14 25 21 28 28 10 22 16 13 8 25 9 11 23 18 7 26 19 3	2.115E-07
30	1 22 20 27 17 29 18 8 15 19 23 14 11 9 26 21 16 13 6 10 4 29 7 24 25 5 3 2 12 28	1.341E-07
31	1 7 28 29 9 13 6 19 2 15 25 10 4 23 11 20 21 18 12 26 5 30 14 16 30 8 24 3 27 17 22	7.939E-08
32	1 18 10 28 17 14 2 16 24 30 9 11 31 5 19 12 6 29 20 31 25 26 23 13 22 3 4 21 15 27 8 7	4.702E-08

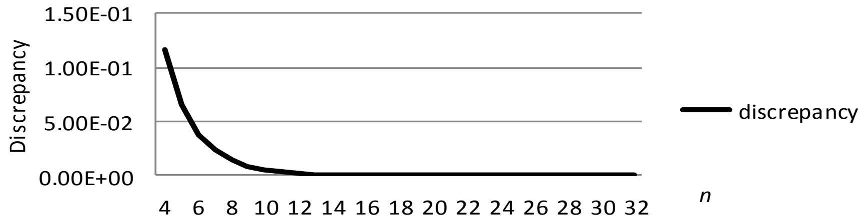


Figure-4.3. L_2-discrepancy against n for DS_1.

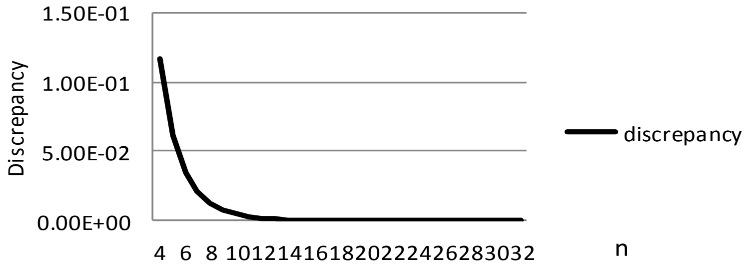


Figure-4.4. L_2-discrepancy against n for DS_2.

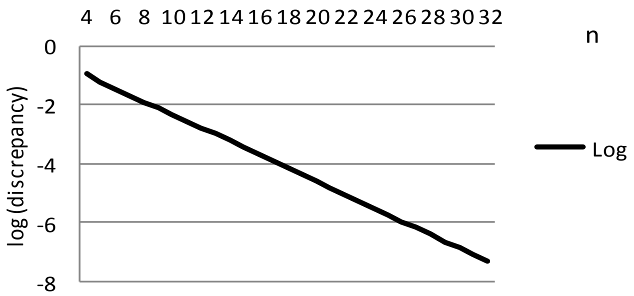


Figure-4.5. Plot of $\log(D_2)$ against n for DS_2.

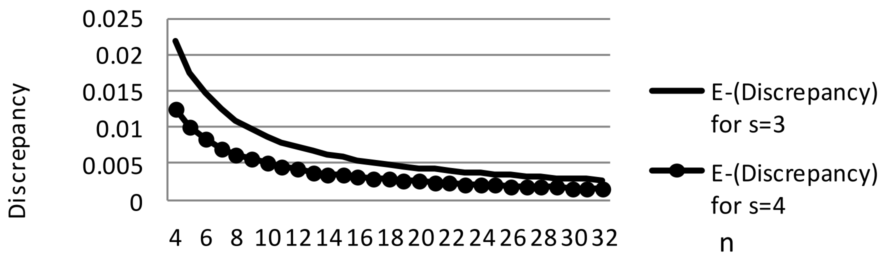


Figure-4.6. $E([D_2(\mathcal{P})]^2)$ against n for s = 3 and 4 for DS_1.

4.5.1 Numerical Comparison between G-Uniform Designs and F-Square Designs

Table 4.6 presents the G-Uniform designs of Fang and Wang (1981) for the particular case $n = 5$ and $s = 4$.

Table 4.6. G-Uniform design for n = 5 and s = 4

No \ Factors	1	2	3	4
1	1	2	4	3
2	2	4	3	1
3	3	1	2	4
4	4	3	1	2
5	5	5	5	5

On comparing the discrepancy of the G-uniform designs and the F-uniform designs for both difference sequences DS_1 and DS_2, we conclude that for $s \geq 2$, the F-uniform designs perform better than the G-uniform designs for many n. Let D_g be the discrepancy of the G-uniform design and D_f be the discrepancy of the F-uniform design for given n and s. The relative improvement in discrepancy is given by $I_1 = (D_g - D_f) / D_g$ or $I_2 = (D_g - D_f) / D_f$. Our results show that I_1 and I_2 range from 0.01% to above 300% for $s \geq 3$, if $D_g > D_f$. For example, for $n = 5$ and $s = 4$, $D_g = 0.3901$, $D_f = 0.083401$, $I_1 = 78\%$ and $I_2 = 367.74\%$.

4.5.2 Numerical Comparison between Latin Square Based Designs and F-Square Designs

Table 4.7 presents the comparison of centered L_2-discrepancy (CD_2) of latin square based designs with F-uniform designs for $n = 4$ to 32. D_L and D_F is the centered L_2-discrepancy of latin square based designs and F-uniform designs for DS_1 and DS_2, respectively. Note that the numerical value of the centered L_2-discrepancy for both the sequences DS_1 and DS_2 is the same. We have made the comparisons till $n = 32$ and have arrived at the conclusion that the centered L_2-discrepancy is lower for F-uniform designs as compared to latin square based designs given by Fang et al. (1999).

Table 4.7. CD_2 of Latin square based design and F-uniform design

n	D_L	D_F
4	0.772814	0.731048
5	0.875065	0.839810
6	0.969241	0.940606
7	1.058232	1.035011
8	1.143819	1.124853
9	1.227226	1.211594
10	1.309349	1.296351
11	1.390883	1.379988
12	1.473391	1.463191
13	1.554340	1.546523
14	1.637134	1.630454
15	1.721127	1.715391
16	1.806638	1.801690
17	1.893958	1.889674
18	1.983359	1.979636
19	2.075097	2.071852
20	2.169414	2.166578
21	2.266547	2.264062
22	2.366724	2.364542
23	2.470171	2.468250
24	2.577110	2.575416
25	2.687761	2.686266
26	2.802347	2.801024
27	2.921091	2.919919
28	3.044217	3.043178
29	3.172738	3.171031
30	3.304535	3.303714
31	3.442197	3.441466
32	3.585182	3.584530

4.6. Wrap-around Discrepancy

The wrap-around L_2-discrepancy proposed by Hickernell (1998b) has the following analytical form

$$\left(WD_2(\mathcal{P})\right)^2 = -\left(\frac{4}{3}\right)^s + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(\frac{3}{2} - |x_{ki} - x_{ji}|\left(1 - |x_{ki} - x_{ji}|\right)\right) \quad (4.11)$$

where $x_k = (x_{k1}, \ldots, x_{ks}) \in \mathcal{P}$.

Table 4.8. Wrap-around discrepancy of latin square and F-square based design

n	WAD_F with DS_1	WAD_F with DS_2	WAD_L
4	3.015327	3.015327	3.153916
5	3.741715	3.736815	3.901141
6	4.665084	4.651127	4.848546
7	5.839945	5.811906	6.040774
8	7.337216	7.277992	7.538449
9	9.248450	9.141039	9.461588
10	11.641603	11.501993	11.879220
11	14.818589	14.503909	14.962099
12	18.825059	18.343177	18.882574
13	23.963053	23.207926	23.865733
14	30.557736	29.331733	30.124196
15	39.026494	37.081422	38.019176
16	49.910190	47.251309	48.409134
17	63.904365	59.738479	61.178003
18	81.906728	76.012810	77.638597
19	105.075526	96.414281	98.493707
20	134.905573	122.427713	124.950040
21	173.326142	155.787366	158.885393
22	222.828572	198.274211	201.980317
23	286.629508	251.430061	255.920323
24	368.884073	320.744031	326.348849
25	474.959743	410.288627	417.094519
26	611.791778	518.371792	526.474678
27	788.342581	659.403505	669.534923
28	1016.196126	854.856595	868.657318
29	1310.327145	1082.363339	1098.554751
30	1690.096557	1352.533725	1371.822194
31	2180.539849	1740.047449	1764.843487
32	2814.034760	2187.659716	2216.637879

WAD_L and WAD_F is the Wrap-around discrepancy of latin square based designs and F-uniform designs for DS_1 and DS_2, respectively. Note that the numerical value of the Wrap-around discrepancy for both the sequences DS_1 and DS_2 is different. The value of Wrap-around discrepancy in the sequences DS_1 like (1 2 3 3) and (1 2 3 1) for n = 4 and (1 2 3 4 4) and (1 2 3 4 1) for n = 5 and all sequences have same discrepancy. The value of the Wrap-around discrepancy for the sequence DS_1 and latin square based designs are greater than the sequence DS_2. The value of the Wrap-around discrepancy for the sequence DS_1 is less than latin square based designs for n = 4 to 12 and is greater for n = 13 to 32, respectively.

4.7. Conclusions

In many cases, the proposed F-uniform designs may significantly improve the uniformity of the corresponding G-uniform designs. The TA algorithm is efficient in locating the "best" LCFS. LCSFS and LCFS whose first rows are given in Tables 4.4 and 4.5 have full rank for both the difference sequences DS_1 and DS_2. Hence for constructing F-uniform designs, we always have n linearly independent columns while there are less alternatives as regards G-uniform designs. Moreover, for the same n and s, we have two difference sequences, namely DS_1 and DS_2 which may be chosen according to the practical requirements. Our results show that indeed, the G-uniform designs have poor uniformity as postulated by Fang et al. (1999). However, as far as centered L_2-discrepancy is concerned, both the sequences DS_1 and DS_2 are equivalent and the centered L_2-discrepancy is lower for F-uniform designs as compared to the latin square based designs of Fang et al. (1999). The value of the Wrap-around discrepancy for the sequence DS_1 and latin square based designs are greater than the sequence DS_2. The value of the Wrap-around discrepancy for the sequence DS_1 is less than latin square based designs for n = 4 to 12 and is greater for n = 13 to 32.

Chapter 5

F-SQUARES BASED EFFICIENT UNIFORM DESIGNS FOR MIXTURE EXPERIMENTS IN THREE AND FOUR COMPONENTS

5.1. Introduction

Uniform design (UD) is a kind of statistical experimental design. It is a kind of space filling design that searches those experimental points that are uniformly scattered over the experimental domain in the sense of low discrepancy (Fang and Wang, 1994). Uniform design (UD) was proposed by Fang (1980), Fang and Wang (1981) and has been popularly used since 1980. One of the several benefits of performing UD is that it helps to study the relationships between the factors and the response of interest using economical number of runs.

In the general mixture experimental setup, the usual constraints on the component proportions are that they are non-negative and should sum to unity. As a result, the factor space reduces to a regular ($q-1$) dimensional simplex

$$S_{q-1} = \left\{x : \left(x_1, x_2, ..., x_q\right) \mid \sum_{i=1}^{q} x_i = 1, x_i \geq 0, i = 1, 2, ..., q\right\} \tag{5.1}$$

In many situations, there may be additional constraints on some or all the factors. For instance, the factors may lie within the lower (L_i) and upper (U_i) bounds

$$0 \leq L_i \leq x_i \leq U_i \leq 1, \qquad i = 1, 2, \ldots, n. \tag{5.2}$$

In such cases, the experimental region is a part of the simplex $S_{q\text{-}1}$. For exploring the restricted region, Mclean and Anderson (1966) have developed extreme vertices designs (EVD) which satisfy both constraints (5.1) and (5.2). Saxena and Nigam (1977) gave a transformation that provides designs constructed through symmetric simplex designs.

Various model forms for mixture experiments are suggested in Chapter 1. The following are the models considered by us in this study.

The quadratic model due to Scheffé (1958):

Model I: $$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} x_i x_j . \tag{5.3}$$

The special cubic model due to Scheffé (1958):

Model II: $$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} x_i x_j + \sum_{1 \leq i < j < k \leq q} \beta_{ijk} x_i x_j x_k . \tag{5.4}$$

The full cubic model due to Scheffé (1958):

Model III:

$$E(Y)=\sum_{i=1}^{q}\beta_i x_i+\sum_{1\le i<j\le q}\beta_{ij}x_i x_j+\sum_{1\le i<j\le q}\beta_{ij}x_i x_j(x_i-x_j)+\sum_{1\le i<j<k\le q}\beta_{ijk}x_i x_j x_k. \tag{5.5}$$

The quadratic additive model due to Darroch and Waller (1985):

Model IV: $$E(Y)=\sum_{i=1}^{q}\beta_i x_i+\sum_{i=1}^{q}\beta_{ii}x_i(1-x_i). \tag{5.6}$$

The homogeneous models of degree one due to Becker (1968)

Model H_i: $$E(Y)=\sum_{i=1}^{q}\beta_i x_i+\sum_{i<j}\beta_{ij}f(x_i,x_j)+\ldots+\sum_{i_1<i_2<\ldots<i_n}\beta_{i_1 i_2\ldots i_n}f(x_i,x_j,\ldots,x_{i_n}), \tag{5.7}$$

where

$$f(x_{i_1},x_{i_2},\ \ldots,x_{i_n})=\min(x_{i_1},\ x_{i_2},\ \ldots,\ x_{i_n}) \qquad \text{for } \textit{Model } H_1$$

$$=\frac{(x_{i_1}x_{i_2}\ldots x_{i_n})}{(x_{i_1}+x_{i_2}+\ldots+x_{i_n})^{n-1}} \qquad \text{for } \textit{Model } H_2$$

$$=(x_{i_1}x_{i_2}\ldots\ x_{i_n})^{1/n} \qquad \text{for } \textit{Model } H_3$$

and $2\le n\le q$.

In *Model H_2* if any denominator is zero, the value of corresponding term is taken to be zero. *Models I*, *II* and *III* are the most commonly used models in mixture experiments. *Model IV* is additive in mixture components and is suitable for the design of industrial or agricultural products where the mixture components have additive effects on the response function. The models introduced by Becker (1968) are homogeneous models of degree one and are applied in different scientific areas. Snee (1979) and Cornell (2002) have described situations such as flare experimental data and strawberry mite experimental data, respectively where these models are applied and found to be better than polynomial models.

Wang and Fang (1990) and Fang and Yang (2000) generated designs for unconstrained and constrained mixture experiments using uniform designs based on good lattice point method. Fang, Shiu and Pan (1999) have defined a new approach based on cyclic latin squares to construct designs on unit hypercube. Box and Hau (2001) and Prescott (2000) have discussed the construction of mixture designs by projecting the standard designs such as two-level factorial and central composite designs. They have also shown that some useful properties of the generating designs such as orthogonal blocking and rotatability are retained in projected designs, which makes these designs suitable for mixture experiments. Prescott (2000) has also discussed the case when some ingredients are restricted to small values.

In the previous chapter, we have obtained uniform designs based on F-squares. In this chapter, we have obtained mixture designs for three and four components by projecting the two families of designs based on good lattice point method. The uniformity measure for these designs is also calculated and tabulated. The D-, A- and G-efficiencies of these designs is also compared. We have also constructed designs for the restricted exploration of mixtures, using the transformation given by Saxena and Nigam (1977). The method has been illustrated with the help of examples.

5.2. Uniform Designs and Uniformity Measures

Uniformity is an important concept in uniform designs. Fang and Wang (1994) described uniform designs (UD) in which the points are scattered uniformly over the experimental domain. This is based on cyclic F-squares. The UD generated by them have smaller discrepancies than those based on the good lattice point method. They have used L_2-discrepancy given by Warnock (1972) as a measure to find the uniform designs.

Warnock (1972) gave the following analytical expression for calculating L_2-discrepancy

$$(D_2(P))^2 = 3^{-s} - \frac{2^{1-s}}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}(1-x_{kl}^2) + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}(1-max(x_{ki}, x_{ji})), \qquad (5.8)$$

where $P = \{x_1, x_2, \ldots, x_n\}$ is a set of n points in $C^s = [0,1]^s$.

Hickernell (1998a) gave an analytical expression for the centered L_2-discrepancy

$$\left(CD_2(P)\right)^2 = \left(\frac{13}{12}\right)^s - \frac{2}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}\left(1+\frac{1}{2}|x_{kl}-0.5|-\frac{1}{2}|x_{kl}-0.5|^2\right)$$
$$+\frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(1+\frac{1}{2}|x_{ki}-0.5|+\frac{1}{2}|x_{ji}-0.5|-\frac{1}{2}|x_{ki}-x_{ji}|\right). \tag{5.9}$$

The centered L_2-discrepancy (CD_2) considers the uniformity of P not only over the unit cube C^s= [0, 1]^s but also of all the projection uniformity of P over C^u where u is a non-empty subset of the set of coordinates indices $\boldsymbol{X} = \{1, 2, \ldots, q\}$.

In this chapter, we have used the centered L_2-discrepancy (CD_2) as a measure of uniformity and the minimum value of CD_2 is desirable for F-square based uniform designs.

5.3. Design Efficiencies Based on Optimality Criteria

Design optimality criteria are often used to evaluate the proposed experimental design for a particular experiment of interest. The following three are the most popular design optimality criteria available in the literature where $\boldsymbol{X}$ denotes the extended design matrix.

- *D*-criterion: maximize the determinant of $|\boldsymbol{X'X}|$
- *A*-criterion: minimize the trace of $(\boldsymbol{X'X})^{-1}$
- *G*-criterion: minimize the maximum of the prediction variance over a specified set of design points.

In order to compare the different designs efficiencies, we use the following most commonly used design optimality measures.

$$D\text{-}eff = 100\left(\frac{|\boldsymbol{X}'\boldsymbol{X}|^{1/p}}{n}\right)$$

$$A\text{-}eff = 100\left(\frac{p}{n\times trace(\boldsymbol{X}'\boldsymbol{X})^{-1}}\right)$$

$$G\text{-}eff = 100\left(\frac{p}{n \times d}\right) \tag{5.10}$$

where,

n = number of design points in the design

p = number of parameters in the model

d = max $\{\nu = \boldsymbol{X}(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\}$ over a specified set of design points (the row vectors) $\boldsymbol{x}$ in $\boldsymbol{X}$.

In this chapter, the efficiencies are generated using Matlab software and are denoted here simply by D, A and G for the sake of convenience. The maximum value of D, A and minimum G are desirable.

5.4. Projection Designs

Prescott (2000) and Box and Hau (2001) have discussed the construction of projection designs for the cases when the design variables are subject to linear constraints. A design satisfying linear constraints has been obtained by projecting unconstrained design onto the constrained space. If q operational factors $x = \{x_i\}$, where $i = 1, 2, \ldots, q$, are subject to m constraints so that

$$\boldsymbol{C}x = \boldsymbol{c} \tag{5.11}$$

where $\boldsymbol{C}$ is an $m \times q$ matrix and $\boldsymbol{c}$ is an $m \times 1$ column vector. Suppose $\boldsymbol{x_0}$ is the chosen origin for the levels of the experimental design then $\boldsymbol{Cx_0} = \boldsymbol{c}$. Let the region of interest be the neighborhood $x_{jo} \pm r_j$ around $\boldsymbol{x_0}$ where r_j's are some positive numbers, then the coded variables $\xi_j = \dfrac{x_j - x_{jo}}{ar_j}$ satisfy the constraints

$$\boldsymbol{A}x = 0, \tag{5.12}$$

where ξ is a $q \times 1$ vector of coded variables ξ_j's, $\boldsymbol{A} = (a_{ij})$ is an $m \times q$ matrix of constraints such that $a_{ij} = r_j c_{ij}$ and 0 is an $m \times 1$ vector of 0's and a is the number to be determined.

Let the $n \times q$ matrix $\boldsymbol{D}_z$ be that of some unconstrained generating design and $\boldsymbol{D}_\xi$ be that of the corresponding constrained design obtained by projection to satisfy (5.12) so that

$$\boldsymbol{D}_\xi = \boldsymbol{D}_z\boldsymbol{P} \tag{5.13}$$

where $\boldsymbol{P}$ is the $q \times q$ idempotent projection matrix

$$\boldsymbol{P} = \boldsymbol{I} - \boldsymbol{R}^T(\boldsymbol{R}\boldsymbol{R}^T)\ \boldsymbol{R} \tag{5.14}$$

Then, as required

$$\boldsymbol{D}_\xi\boldsymbol{A}^T = \boldsymbol{D}_z\boldsymbol{P}\ \boldsymbol{A}^T = \boldsymbol{0} \tag{5.15}$$

and the levels of the design $\boldsymbol{D}_X$ may be obtained from

$$x_j = ar_j\xi_j + x_{j0} \tag{5.16}$$

where 'a' is the number such that all the entries of $a\boldsymbol{D}_\xi$ are between -1 and 1.

5.5. Unconstrained Mixture Experiments

Aggarwal and Singh (2008) suggested a method to construct latin square based design of n runs for mixture of q components. We describe the following method to construct mixture designs through cyclic F-squares. UF-type designs may be obtained by selecting s linearly independent columns of a cyclic F-square given in Chapter 4, Section 4.5.

Method

To construct F-squares based n run mixture design in q components for UF-type designs.

Step-1: Choose a *UF*-type design $UF(n; q^n)$ and any $s(\leq n)$ columns of an F-square to form a *UF*-type design $UF(n; q^s)$.

Step-2: For a given n, there are n! left cyclic F-squares. Among all these n! left cyclic F-squares of order n, find an F-square $F = f_{ij}$ that has the smallest discrepancy using the expression given in (5.9).

Step-3: Search $s = (q\text{-}1)$ out of n columns of the F-square to form a *UF*-type design $UF(n; q^s)$. In all there are ${}^nC_s$ such *UF*-type designs.

Step-4: From these ${}^nC_s$ *UF*-type designs $UF(n; q^s)$, choose a design $UF_n(n^s)$ such that it has the smallest discrepancy (as given in (5.9)) among all $UF(n; q^s)$ designs generated in Step-3. This design is nearly uniform design. We now have $UF_n(n^s)$ on C^s where $C = [0,1]$

Step-5: Let $U = (u_{ki})$, $u_{ki} = kh_i \pmod n$, $i = 1, 2, \dots, s$; $k = 1, 2, \dots, n$ be the uniform design as obtained in Step-4. Calculate $C_{ki} = (u_{ki} - 0.5) / n$ and make the transformation given in Fang and Wang (1994, p.231).

$$x_{ki} = \prod_{j=1}^{i-1} C_{kj}^{\frac{1}{q-j}} \left(1 - C_{ki}^{\frac{1}{q-i}}\right), \qquad i = 1, 2, \dots, q\text{-}1.$$

$$x_{kq} = \prod_{j=1}^{q-1} C_{kj}^{\frac{1}{q-j}}, \qquad k = 1, 2, \dots, n. \tag{5.17}$$

then $x_k = (x_{k1}, x_{k2}, \dots, x_{kq})$, $k = 1, 2, \dots, n$; is a uniform design on $S_{q\text{-}1}$.

Note that, the number of possible h_i is given by the Euler function $\varphi(n)$ defined in (5.18) by Hua (1956) as follows:

$$\varphi(n) = n \prod_{p|n} \left(1 - \frac{1}{p}\right), \tag{5.18}$$

where p runs over the prime divisors of n. For example,

$$\varphi(6) = 6\left(1 - \frac{1}{2}\right)\left(1 - \frac{1}{3}\right) = 2$$

because $6 = 2 \times 3$, and the possible associated h_i with $(h_i, n) = 1$ are 1 and 5.

In this chapter, we have presented two classes of uniform designs for mixture application. We have studied properties of these classes of designs for various models. The two classes of considered designs are (i) mixture designs constructed using uniform designs based on cyclic F-squares for two designs with different runs, (ii) mixture designs constructed by projecting the uniform designs based on good lattice point method based on the two designs with different runs. Consider the case n = 4. We have taken 4 = 3 as it yields centered L_2-discrepancy (CD_2), the natural order being 1 2 3 3. This sequence is denoted with last-previous (*LP*) value. For the other sequence, we have taken 4 = 1 as it yields centered L_2-discrepancy (CD_2), the natural order being 1 2 3 1. We have denoted this sequence with last-first (*LF*) value. Hence the considered designs with different runs are denoted by *LP* and *LF*.

Table 5.1: Discrepancies of the most uniform three component mixture designs

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.672528	0.691977	0.621585	0.626328
5	0.680799	0.703155	0.628167	0.632778
6	0.685391	0.705232	0.631856	0.636886
7	0.690831	0.703638	0.628626	0.631139
8	0.702989	0.711591	0.641498	0.644163
9	0.698299	0.708262	0.641358	0.644810
10	0.699276	0.710290	0.633579	0.636435
11	0.688570	0.709356	0.634094	0.636934
12	0.708748	0.716759	0.639239	0.641616
13	0.705680	0.714011	0.637435	0.640063
14	0.691894	0.698163	0.634056	0.635911
15	0.702877	0.702108	0.649156	0.648736
16	0.711894	0.716383	0.645628	0.647331
17	0.707727	0.708599	0.652365	0.652775
18	0.707581	0.708261	0.652794	0.653077
19	0.712794	0.717675	0.643690	0.645387
20	0.707428	0.707763	0.648146	0.653689

Using the method given above, we have first obtained the most uniform designs based on cyclic F-squares. These designs are denoted here by D_{UF}. Then using step 5, we have obtained mixture designs for three and four component mixtures. These are denoted here by D_F. We have also generated mixture designs through projection of uniform designs D_{UF} as described in Section 5.4. So we now have two classes of designs D_F and D_{UF} for both the sequences *LP* and *LF*, respectively. The uniformity measure for each of these classes of designs is calculated using (5.9). The discrepancies CD_2 for each of the classes of mixture designs in three and four components are given in Table 5.1 and Table 5.2. The most uniform six run designs for three and four component mixtures for each of the families D_F and D_{UF} are given in Table 5.3 and Table 5.4, respectively.

Table 5.2: Discrepancies of the most uniform four component mixture designs

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.861168	0.869273	0.827342	0.824991
5	0.865524	0.875347	0.825065	0.825174
6	0.865774	0.878883	0.825161	0.825317
7	0.866703	0.878016	0.825020	0.824307
8	0.874400	0.877889	0.825441	0.825482
9	0.877626	0.884636	0.825589	0.825948
10	0.875257	0.880175	0.825236	0.825302
11	0.874308	0.879774	0.825245	0.825308
12	0.882991	0.887693	0.825711	0.825810
13	0.876571	0.881749	0.825347	0.825432
14	0.873899	0.874391	0.825328	0.825329
15	0.884629	0.886514	0.826045	0.826092
16	0.886045	0.888752	0.826175	0.826248
17	0.885668	0.889178	0.826141	0.826252
18	0.874407	0.877318	0.825825	0.825917
19	0.876212	1.745720	0.825630	0.825706
20	0.872871	0.874093	0.825864	0.825909

From Table 5.1, we observe that for three component mixtures, the design $D_{UF}(LF)$ is most uniform for run sizes $n = 15$ for the two classes D_F and D_{UF} and $D_{UF}(LP)$ is most uniform for all run sizes expect $n = 15$. All designs are most uniform for $n = 4$ runs. From Table 5.2, we observe that for four component mixtures, the design $D_{UF}(LF)$ is most uniform for run sizes $n = 4$ and 7 and the design $D_{UF}(LP)$ is most uniform for all run sizes expect $n = 4$ and 7. When $n = 4$, the designs $D_F(LP)$ and $D_F(LF)$ are most uniform. When $n = 7$, the designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are most uniform. The designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are obtained through projection of the designs $D_F(LP)$ and $D_F(LF)$. The designs $D_F(LP)$ and $D_{UF}(LF)$ are most uniform better than the designs $D_F(LF)$ and $D_{UF}(LP)$, respectively.

Table 5.3: The most uniform six run designs for three component mixtures

$D_F(LP)$			$D_F(LF)$		
0.7113	0.2165	0.0722	0.7113	0.2165	0.0722
0.2362	0.4455	0.3182	0.2362	0.4455	0.3182
0.5000	0.1250	0.3750	0.5000	0.1250	0.3750
0.3545	0.1614	0.4841	0.3545	0.5917	0.0538
0.1340	0.7939	0.0722	0.1340	0.7939	0.0722
0.1340	0.3608	0.5052	0.7113	0.1203	0.1684
$D_{UF}(LP)$			$D_{UF}(LF)$		
0.5074	0.2795	0.2131	0.5074	0.2795	0.2131
0.2886	0.3850	0.3264	0.2886	0.3850	0.3264
0.4101	0.2374	0.3525	0.4101	0.2374	0.3525
0.3431	0.2542	0.4028	0.3431	0.4523	0.2046
0.2415	0.5454	0.2131	0.2415	0.5454	0.2131
0. 2415	0.3460	0.4125	0. 5074	0.2352	0.2574

Table 5.4: The most uniform six run designs for four component mixtures

$D_F(LP)$				$D_F(LF)$			
0.5632	0.2184	0.0546	0.1638	0.5632	0.2184	0.0546	0.1638
0.1645	0.2962	0.1348	0.4045	0.1645	0.2962	0.4944	0.0449
0.3700	0.0844	0.5001	0.0455	0.3700	0.0844	0.5001	0.0455
0.2531	0.1001	0.2695	0.3773	0.2531	0.5313	0.0898	0.1258
0.0914	0.6463	0.1967	0.0656	0.0914	0.6463	0.1967	0.0656
0.0914	0.2146	0.4048	0.2891	0.5632	0.1032	0.1946	0.1390
$D_{UF}(LP)$				$D_{UF}(LF)$			
0.2810	0.2469	0.2306	0.2415	0.2810	0.2469	0.2306	0.2415
0.2415	0.2546	0.2386	0.2653	0.2415	0.2546	0.2742	0.2297
0.2619	0.2336	0.2748	0.2297	0.2619	0.2336	0.2748	0.2297
0.2503	0.2351	0.2519	0.2626	0.2503	0.2779	0.2341	0.2377
0.2343	0.2893	0.2447	0.2317	0.2343	0.2893	0.2447	0.2317
0.2343	0.2465	0.2653	0.2539	0.2810	0.2355	0.2445	0.2390

We have fitted *Model I* to *Model IV* to the minimum point most uniform mixture designs in three and four components for each of the two classes. *Models Hi*; i = 1, 2, 3 to the minimum point most uniform mixture designs in three component and *Models*

Hi; i = 2, 3 to the minimum point most uniform mixture designs in four components for each of the two classes *LP* and *LF*, respectively . The design efficiencies D, A and G are given in Table 5.5 and Table 5.6, respectively.

From Table 5.5, we observe that for three component mixtures, the designs generated from $D_F(LP)$ and $D_F(LF)$ are in general more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$ for all *Model I* to *IV* and *Models Hi*; i = 1, 2, 3 as regards D-efficiency. As regards A-efficiency, for *Model II* to *IV*, the designs generated from $D_F(LP)$ and $D_F(LF)$ are more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$ and in other models, the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$ are more efficient than the designs generated from $D_F(LP)$ and $D_F(LF)$. From Table 5.6, we observe that the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$ are in general more efficient than the designs generated from $D_F(LP)$ and $D_F(LF)$. Designs based on $D_{UF}(LP)$ is better in terms of efficiencies for *Model I*, *III* and H_2. Designs based on $D_{UF}(LF)$ is better in terms of efficiencies for *Model II*. Designs based on $D_F(LP)$ are better for *Model IV* in terms of D-efficiency.

5.6. Restricted Exploration of Mixtures

For restricted exploration of mixtures i.e., when (5.2) is satisfied, Saxena and Nigam (1977) have given a transformation which provides designs constructed through symmetric simplex designs. Aggarwal and Singh (2008) suggested the following steps to generate projection designs based on Uniform designs.

Step 1: Rank the components in order of their increasing ranges ($U_i - L_i$). x_1 has the smallest range and x_q has the largest range.

Step 2: Consider a mixture design Z satisfying (5.1). This can be selected from the two classes of designs obtained in Section 5.5.

Step 3: Compute B and B', the minimum and maximum proportions of any component Z_i in the design so that $0 \le B \le Z_i \le B' \le 1$ for all Z_i.

Step 4: Make the transformation as given by Saxena and Nigam (1977) i.e.,

$$x_{iu} = \lambda_i + \mu_i z_{iu}, \qquad i = 1, 2, \ldots, t; \;\; u = 1, 2, \ldots, n$$

where

$$\lambda_i = \frac{L_i B' - U_i B}{B' - B} \qquad \textit{and} \qquad \mu_i = \frac{U_i - L_i}{B' - B}$$

and

$$x_{iu} = \frac{\left(1 - \sum_{h=1}^{t} x_{hu}\right)}{\left(1 - \sum_{h=1}^{t} z_{hu}\right)} z_{iu} \qquad i = t+1, \ldots, q; u = 1, 2, \ldots, n$$

where $t \leq (q\text{-}1)$ is the number of components constrained by (5.2).

When all the components are constrained by (5.2), then the levels of x_q may be obtained by $x_q = 1 - (x_1 + x_2 + \cdots + x_{q-1})$.

Step-5: While determining the value of x_q in Step-4, if at any point, x_q lie outside the range $L_q \leq x_q \leq U_q$, it can be adjusted by setting x_q equal to the violated bound and adjusting the level of x_{q-1} so that (5.2) is satisfied.

Step-6: The design points from Step-4 combined with different combinations of adjusted points result in a number of designs. The design that is most uniform and optimal with certain optimality criteria is taken as the best design.

The steps given above are illustrated with the help of examples for three and four component mixtures based on F-squares.

Example 5.1: Let us first consider a three component example taken from Snee and Marquardt (1974) with components ranked in order of their increasing ranges.

$$0.1 \leq x_1 \leq 0.6$$

$$0.1 \leq x_2 \leq 0.7$$

$$0.0 \leq x_3 \leq 0.7$$

Table 5.5: Efficiencies of the minimum point uniform mixture designs for three components

Model	*p*	$D_F(LP)$			$D_F(LF)$			$D_{UF}(LP)$			$D_{UF}(LF)$		
		D-eff	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*
I	6	9.7375E-06	1.2782E-15	1.5E-12	4.42975E-06	1.4154E-16	1.5E-14	3.4636E-06	3.8078E-15	1.5E-13	2.552E-06	7.8113E-16	1.5E-12
II	7	1.8645E-07	1.7255E-16	1.75E-13	1.1053E-07	3.2596E-18	1.75E-13	8.4310E-08	1.3334E-16	1.75E-12	5.5437E-08	3.7433E-17	1.75E-13
III	10	2.4114E-10	4.3806E-17	2.5E-14	2.4021E-10	1.1599E-14	2.5E-14	9.7349E-11	2.7202E-17	2.5E-13	1.1219E-10	6.2470E-18	2.5E-13
IV	6	8.6172E-06	8.9648E-15	1.5E-13	1.2528E-05	8.6331E-16	1.5E-13	2.8269E-06	1.1255E-15	1.5E-13	4.5387E-06	1.1724E-14	1.5E-13
$H_1(r=3)$	7	4.3705E-07	1.4747E-16	1.675E-13	2.4378E-07	4.6357E-17	1.675E-13	2.9958E-07	7.4569E-16	1.675E-12	2.7274E-07	1.8211E-14	1.675E-14
$H_2(r=3)$	7	2.4565E-07	1.1828E-16	1.675E-12	1.2875E-07	8.4545E-17	1.675E-12	7.3837E-08	7.8328E-16	1.675E-11	5.1464E-08	1.0229E-16	1.675E-12
$H_3(r=3)$	7	6.1651E-07	1.2793E-15	1.675E-12	5.2911E-07	7.5107E-16	1.675E-13	1.1305E-07	2.6181E-15	1.675E-12	2.7114E-07	2.1387E-15	1.675E-12

Table 5.6: Efficiencies of the minimum point uniform mixture designs for four components

Model	*p*	$D_F(LP)$			$D_F(LF)$			$D_{UF}(LP)$			$D_{UF}(LF)$		
		D-eff	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*
I	10	4.9095E-07	3.9135E-16	1.6667E-13	2.7296E-07	8.2451E-16	1.6667E-13	9.054E-07	2.2291E-16	1.6667E-13	1.0433E-08	7.2376E-16	1.6667E-13
II	14	1.7115E-12	7.1987E-16	3.5E-14	1.3150E-12	7.1987E-16	3.5E-14	1.8794E-10	4.1135E-16	2E-13	2.7359E-11	4.1135E-16	2E-14
III	20	4.9714E-14	1.0284E-15	5E-14	3.6525E-14	1.0284E-15	5E-14	9.1968E-13	5.8765E-16	2.8571E-13	1.7064E-13	2.7822E-19	3.3333E-14
IV	8	0.00016838	2.1635E-16	1.3333E-14	3.7762E-08	4.1135E-16	2E-13	2.3483E-06	3.0953E-16	1.3333E-13	2.3588E-05	2.3506E-16	1.1428E-05
$H_2(r=3)$	14	4.2878E-12	7.1987E-16	3.5E-13	3.2031E-10	7.1987E-16	3.5E-13	3.5250E-10	4.1135E-16	4.1135E-16	6.3163E-11	2.691E-15	2.3333E-13
$H_3(r=3)$	14	4.8407E-09	7.1987E-16	2.3333E-13	2.2733E-09	1.3990E-15	2.3333E-13	3.7487E-10	4.1135E-16	2.3333E-13	2.6706E-10	4.1135E-16	2E-11

Using Steps 1 to 4, we obtain designs with different run sizes for each of the two considered classes. The discrepancies of these designs are calculated for different run sizes and are given in Table 5.7. Table 5.8 presents the six run uniform designs based on designs given in Table 5.3.

From Table 5.7, we observe that for Example 1 when n = 4, 6, 10 and 11, $D_{UF}(LP)$ is most uniform and $D_{UF}(LF)$ is most uniform for n = 5, 7, 8, 9, 12, 13, 14, 15, 16, 17, 18, 19 and 20. For n = 6, 7, 10 and 11, $D_F(LP)$ is most uniform and $D_F(LF)$ is most uniform for n = 4, 5, 8, 9, 12, 13, 14, 15, 16, 17, 18, 19 and 20.

We have fitted *Model I* to *Model IV* and *Models Hi*; i = 1, 2, 3 to the minimum point most uniform mixture designs in three components for each of the four classes. The design efficiencies D, A and G are given in Table 5.11.

Example 5.2: Let us now consider a four component example taken from Snee (1975) with components ranked in order of their increasing ranges.

$$0.07 \leq x_1 \leq 0.18$$
$$0.00 \leq x_2 \leq 0.15$$
$$0.00 \leq x_3 \leq 0.30$$
$$0.37 \leq x_4 \leq 0.70$$

Using Steps 1 to 4, we obtain designs with different run sizes for each of the four classes. The discrepancies of these designs are calculated for different run sizes and are given in Table 5.9. Table 5.10 presents the six run uniform designs based on designs given in Table 5.4.

From Table 5.9, we observe that for Example 5.2 when n = 4, the designs $D_F(LF)$ and $D_{UF}(LF)$ are most uniform. For n = 6, the designs $D_F(LP)$ and $D_{UF}(LP)$ are most uniform. We also observe that for four component mixture designs D_F and D_{UF} with this example are most uniform for all runs as compared to Aggarwal and Singh (2008).

We have fitted *Model I* to *Model IV* and *Models Hi*; i = 2 and 3 to the minimum point most uniform mixture designs in four components for each of the four classes. The design efficiencies D, A and G are given in Table 5.12.

Table 5.7: Discrepancies of most uniform three component designs for Example 5.1

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.681488	0.672802	0.616094	0.616212
5	0.680474	0.660865	0.616276	0.615941
6	0.659181	0.660720	0.615957	0.616017
7	0.664419	0.666565	0.615895	0.615682
8	0.666437	0.658828	0.616133	0.615996
9	0.662670	0.654420	0.616009	0.615910
10	0.661016	0.663801	0.615732	0.615798
11	0.662063	0.672179	0.615694	0.615697
12	0.669371	0.666703	0.616244	0.616067
13	0.666600	0.664524	0.615912	0.615806
14	0.653625	0.652520	0.615572	0.615567
15	0.657551	0.652834	0.616093	0.615860
16	0.665891	0.663251	0.616098	0.616029
17	0.662207	0.655734	0.616225	0.615988
18	0.662159	0.655818	0.616187	0.615928
19	0.667399	0.665513	0.616057	0.616043
20	0.660461	0.656061	0.616198	0.616054

Table 5.8: The most uniform six run designs for three component mixtures

$D_F(LP)$			$D_F(LF)$		
0.5409	0.2332	0.2259	0.5417	0.2393	0.2190
0.2242	0.4164	0.3594	0.2291	0.4202	0.3507
0.4000	0.1600	0.4400	0.4026	0.1671	0.4303
0.3030	0.1891	0.5079	0.3069	0.5356	0.1575
0.1560	0.6951	0.1489	0.1618	0.6951	0.1430
0.1560	0.3487	0.4953	0.5417	0.1634	0.2950
$D_{UF}(LP)$			$D_{UF}(LF)$		
0.3584	0.3213	0.3204	0.3585	0.3220	0.3195
0.3202	0.3434	0.3365	0.3208	0.3438	0.3354
0.5414	0.3124	0.3462	0.3417	0.3133	0.3450
0.3297	0.3159	0.3544	0.3301	0.3577	0.3121
0.3119	0.3770	0.3111	0.3127	0.3770	0.3104
0.3119	0.3352	0.3529	0. 3585	0.3128	0.3287

Table 5.9: Discrepancies of most uniform four component designs for Example 5.2

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.934520	0.905616	0.827773	0.826239
5	0.923403	0.912608	0.827227	0.826572
6	0.917171	0.918592	0.826819	0.826915
7	0.918128	0.912089	0.827080	0.826632
8	0.927806	0.922770	0.827196	0.827245
9	0.937198	0.929950	0.828361	0.827862
10	0.924913	0.920287	0.827630	0.827409
11	0.923215	0.918763	0.827208	0.826723
12	0.933938	0.929168	0.828241	0.827850
13	0.922307	0.919754	0.827224	0.827026
14	0.929262	0.924009	0.827877	0.827445
15	0.934432	0.931890	0.828199	0.827787
16	0.938489	0.934687	0.828649	0.828062
17	0.934953	0.930937	0.828237	0.829316
18	0.933185	0.930975	0.827793	0.827632
19	0.930816	0.932483	0.827756	0.827501
20	0.934301	0.930737	0.827856	0.827671

Table 5.10: The most uniform six run designs for four component mixtures

$D_F(LP)$				$D_F(LF)$			
0.1788	0.0503	0.0075	0.7634	0.1643	0.0437	0.0066	0.7854
0.0956	0.0724	0.0531	0.7788	0.0923	0.0628	0.2233	0.6216
0.1385	0.0122	0.2609	0.5884	0.1294	0.0106	0.2261	0.6338
0.1141	0.0167	0.1297	0.7395	0.1083	0.1208	0.0240	0.7470
0.0804	0.1720	0.0883	0.6593	0.0791	0.1491	0.0766	0.6952
0.0804	0.0492	0.2066	0.6637	0.1643	0.0153	0.0756	0.7448
$D_{UF}(LP)$				**$D_{UF}(LF)$**			
0.2406	0.2236	0.2179	0.3179	0.2385	0.2224	0.2174	0.3216
0.2296	0.2265	0.2240	0.3199	0.2289	0.2250	0.2464	0.2997
0.2353	0.2186	0.2514	0.2947	0.2339	0.2180	0.2468	0.3014
0.2320	0.2192	0.2341	0.3147	0.2310	0.2327	0.2198	0.3165
0.2276	0.2397	0.2286	0.3041	0.2271	0.2365	0.2268	0.3096
0.2276	0.2235	0.2443	0.3047	0.2385	0.2186	0.2267	0.3162

Table 5.11: Efficiencies of the minimum point uniform mixture designs for three components in example 5.1

Model	*p*	$D_F(LP)$			$D_F(LF)$			$D_{UF}(LP)$			$D_{UF}(LF)$		
		D-eff	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*
I	6	0.16350	0.00110	46.7720	0.1861	0.00323	49.7471	0.411E-3	0.43E-6	49.8629	0.324E-3	0.422E-6	53.1726
II	7	0.04740	0.162E-4	54.4781	0.0471	0.742E-4	58.0450	0.742E-4	0.362E-7	53.2255	0.766E-4	0.7694E-7	54.3951
III	10	0.00440	0.716E-5	71.4357	0.0043	0.702E-5	71.4357	0.519E-5	0.114E-7	78.7352	0.522E-5	0.110E-7	67.4554
IV	6	0.20561	0.00243	46.7057	0.2342	0.00784	49.7125	0.00032	0.346E-6	49.0637	0.361E-4	0.556E-6	53.9016
$H_1(r=3)$	7	0.41792	0.01543	58.1531	0.4830	0.04021	52.4329	0.00611	0.882E-4	58.1666	0.00712	0.255E-4	52.5045
$H_2(r=3)$	7	0.04593	0.521E-4	54.9390	0.0436	0.101E-4	54.0482	0.561E-4	0.414E-7	53.8039	0.702E-4	0.205E-7	53.9084
$H_3(r=3)$	7	0.06651	0.196E-4	54.2299	0.0622	0.439E-4	56.1987	0.892E-4	0.783E-7	260.8242	0.961E-4	0.921E-7	50.3221

Table 5.12: Efficiencies of the minimum point uniform mixture designs for four components in example 5.2

Model	*p*	$D_F(LP)$			$D_F(LF)$			$D_{UF}(LP)$			$D_{UF}(LF)$		
		D-eff	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*	*D-eff*	*A-eff*	*G-eff*
I	10	4.47176E-08	1.03244E-17	1.66667E-14	9.64023E-11	3.56379E-17	2.5E-13	7.6344E-09	1.38819E-15	1.6667E-12	5.58969E-11	3.34685E-16	2.5E-13
II	14	2.34133E-11	1.70428E-19	2.33333E-13	4.19051E-13	1.43056E-18	3.5E-14	2.23283E-11	1.27588E-17	2.3333E-13	5.7071E-13	1.55039E-17	3.5E-13
III	20	2.55634E-13	2.19775E-19	3.33333E-15	1.64889E-13	5.17507E-18	5.0E-14	5.19251E-13	5.9459E-18	3.3333E-13	2.26439E-14	3.49895E-20	5E-14
IV	8	1.1314E-05	6.24132E-16	1.33333E-13	1.00699E-08	4.02325E-16	2.0E-14	2.47232E-06	4.85838E-15	1.3333E-12	4.32156E-09	1.81587E-15	2E-13
$H_2(r=3)$	14	9.44261E-11	2.94449E-18	2.33333E-14	1.15283E-12	3.83852E-18	3.5E-13	1.70278E-10	9.13099E-16	2.3333E-13	1.55869E-12	4.89052E-17	3.5E-13
$H_3(r=3)$	14	9.68000E-10	2.03553E-16	2.33333E-14	1.35677E-11	7.30628E-16	3.5E-13	1.83134E-10	2.002E-16	2.3333E-15	1.00185E-11	6.37616E-15	3.5E-13

From Table 5.11, we observe that the designs generated from $D_F(LP)$ is better in terms of D- efficiency for *Model II*, *III*, H_2 and H_3 while $D_F(LF)$ is better for *Model I*, *IV* and H_1. The designs generated from $D_F(LP)$ is better in terms of A- efficiency for *Model III* and H_2 and $D_F(LF)$ is better for *Model I*, *II*, *IV*, H_1 and H_3. The designs generated from $D_F(LP)$ are better in terms of G-efficiency for *Model* H_2, $D_F(LF)$ is better for *Model II*, $D_{UF}(LP)$ is better for *Model III*, H_1 and H_3 while $D_{UF}(LF)$ is better for *Model I* and *IV*.

From Table 5.12, we observe that the designs generated from $D_F(LP)$ and $D_F(LF)$ are in general more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$. Designs based on $D_{UF}(LP)$ is better in terms of efficiencies for *Model III* and H_2. Designs based on $D_F(LP)$ is better in terms of efficiencies for *Model I*, *II*, *IV* and H_3. Designs based on $D_{UF}(LP)$ is better for *Model III* and H_2 as regards D-efficiency.

5.7. Conclusions

For three and four components, D-, A- and G-efficient uniform designs for mixture experiments based on latin squares for Scheffė's quadratic model (1958), Darroch and Waller's (1985) model and Becker's (1968) model were obtained by Aggarwal and Singh (2008). In this chapter, we have found the centered L_2-discrepancy with three and four components for the considered classes of designs based on F-squares. The design $D_{UF}(LF)$ is most uniform for run sizes n = 4 and 7 in three components. When n = 4, all the designs are most uniform for three components. The designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are obtained through projection of the designs $D_F(LP)$ and $D_F(LF)$ based on four components. The designs $D_F(LP)$ and $D_{UF}(LF)$ are most uniform better than the designs $D_F(LF)$ and $D_{UF}(LP)$ for four components.

In this chapter, we have computed the D-, A- and G- efficiencies of three and four component mixture experiments based on F-squares for Scheffė's (1958) quadratic model, Darroch and Waller's (1985) model and Becker's (1968) model. For three components, the designs generated from $D_F(LP)$ is better in terms of D- efficiency for *Model II*, *III*, H_2 and H_3 and $D_F(LF)$ is better in terms of D- efficiency for *Model I*, *IV* and H_1. The designs generated from $D_F(LP)$ is better in terms of A- efficiency for *Model III* and H_2 and $D_F(LF)$ is better for *Model I*, *II*, *IV*, H_1 and H_3. The designs generated from $D_F(LP)$ is better in terms of G- efficiency for *Model* H_2, $D_F(LF)$ is better for *Model II*, $D_{UF}(LP)$ is better for *Model III*, H_1 and H_3 and $D_{UF}(LF)$ is better for *Model I* and *IV*. For four components, the designs generated from $D_F(LP)$ and $D_F(LF)$ are in general more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$. Designs based on $D_{UF}(LP)$ is better in terms of D- efficiency for *Model III* and H_2. Designs based on $D_F(LP)$ is better in terms of D- efficiency for *Model I*, *II*, *IV* and H_3. Design based on $D_{UF}(LP)$ is better for *Model III* and H_2 as regards D-efficiency.

For four components, the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$ are in general more efficient than the designs generated from $D_F(LP)$ and $D_F(LF)$. For Snee's (1975) constrained experimental data, we have compared the discrepancies of our designs with latin square based design obtained by Aggarwal and Singh (2008).

APPENDIX: C++ PROGRAMS FOR CHAPTER 4 AND CHAPTER 5

We have used two methods to obtain the desired discrepancies. In the first method, we have provided the data matrix and in the second method we have just provided the first row of the matrix and the remaining rows are generated by the program itself.

Chapter 4

//Program to find out the L_2 – Discrepancy with matrix input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
        char ch='y';    P
        double m[60][60];
        long k,l,s,i,n,g[60][60];
        double p,sum;
        while(ch=='y' || ch=='Y')
        {
                clrscr();
                cout<<"Enter No. Of Elements n ="; cin>>n;
                cout<<"Enter No. Of Elements s ="; cin>>s;
               cout<<"Enter Matrix Data\n";
                for(k=0;k<n;k++)
                {
                        for(l=0;l<s;l++)
                        {
                                cin>>g[k][l];
                        }
               }
                cout<<" Matrix Data\n";
                for(k=0;k<n;k++)
               {
```

```
            for(l=0;l<s;l++)
            {
                    m[k][l]=(g[k][l]-0.5)/n;
                    cout<<m[k][l]<<"  ";
            }
            cout<<endl;
    }
    sum=0.0;
    for(k=0;k<n;k++)
    {
            p=1.0;
            for(l=0;l<s;l++)
            {
                    p=p*(1-pow(m[k][l],2));
            }
            sum=sum+p;
    }
    cout<<"\nsum of middle value="<<sum;
    double res=pow(3,-s)-((pow(2,(1-s))/n)*sum);
    cout<<"\nresult of first two part="<<res;
    double y=0.0,x;
    double m1;
    for(k=0;k<n;k++)
    {
            x=0.0;
            for(l=0;l<n;l++)
            {
                    p=1.0;
                    for(i=0;i<s;i++)
                    {
                            if(m[k][i]>m[l][i])
                                    m1=m[k][i];
                            else
                                    m1=m[l][i];
```

```
                        p=p*(1.0-m1);
                    }
                    x=x+p;
                }
                y=y+x;
            }
            cout<<"\nresult  y="<<y;
            double res1=res+(y/(n*n));
            cout<<"\nresult total="<<res1;
            cout<<"\nresult total as per square root="<<sqrt(res1);
            cout<<"\nCheck for another(Y/N)=";
            cin>>ch;
    }
}
```

Output

```
Enter No. Of Elements n =5
Enter No. Of Elements s =5
Enter Matrix Data
1 2 3 4 4
2 3 4 4 1
3 4 4 1 2
4 4 1 2 3
4 1 2 3 4
 Matrix Data
0.1  0.3  0.5  0.7  0.7
0.3  0.5  0.7  0.7  0.1
0.5  0.7  0.7  0.1  0.3
0.7  0.7  0.1  0.3  0.5
0.7  0.1  0.3  0.5  0.7

sum of middle value=0.878715
result of first two part=-0.006869
result  y=0.27675
result total=0.004201
result total as per square root=0.064817
Check for another(Y/N)=_
```

// Program to find out the L_2 - discrepancy with the first row input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
        char ch='y';
         double p,sum,m[60][60];
         long k,l,s,i,n,g[60][60],j;
         while(ch=='y' || ch=='Y')
        {
                clrscr();
                cout<<"Enter No. Of Elements=";
                cin>>n;
                 s=n;
                 cout<<"\nEnter 1st Row Matrix Data\n";
                 for(l=0,k=0;l<n;l++)
                {
                        cin>>g[k][l];
                }
                 for(k=1;k<n;k++)
                {
                        for(l=0;l<n-k;l++)
                        {
                                g[k][l]=g[0][l+k];
                        }
                        for(j=0;l<n;l++,j++)
                        {
                                g[k][l]=g[0][j];
                        }
                }
                clrscr();
```

```
 cout<<" Matrix Data\n";
 for(k=0;k<n;k++)
 {
         for(l=0;l<n;l++)
        {
                cout<<g[k][l]<<" ";
        }
        cout<<endl;
}
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                m[k][l]=(g[k][l]-0.5)/n;
        }
}
 sum=0.0;
 for(k=0;k<n;k++)
{
        p=1.0;
        for(l=0;l<n;l++)
        {
               p=p*(1-pow(m[k][l],2));
        }
        sum=sum+p;
}
double res=pow(3,-s)-((pow(2,(1-s))/n)*sum);
double y=0.0;
 double m1;
 for(k=0;k<n;k++)
{
         double x=0.0;
        for(l=0;l<n;l++)
        {
```

```
                    p=1.0;
                    for(i=0;i<n;i++)
                    {
                        if(m[k][i]>m[l][i])
                            m1=m[k][i];
                        else
                            m1=m[l][i];
                        p=p*(1.0-m1);
                    }
                    x=x+p;
                }
                y=y+x;
            }
            double res1=res+(y/(n*n));
            cout<<"result total="<<res1;
            cout<<"\nresult total as per square root="<<sqrt(res1);
            cout<<"\nCheck for another(Y/N)=";
            cin>>ch;
    }
}
```

Output

```
 Matrix Data
1 2 3 4 5 5
2 3 4 5 5 1
3 4 5 5 1 2
4 5 5 1 2 3
5 5 1 2 3 4
5 1 2 3 4 5
result total=0.001455
result total as per square root=0.038149
Check for another(Y/N)=
```

// program to find the Centered L_2 discrepancy with matrix input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
	char ch='y';
	 double p,sum,m[60][60];
	 long k,l,s,i,n,g[60][60],j;
	 while(ch=='y' || ch=='Y')
	{
		clrscr();
		cout<<"Enter No. Of Elements n=";
		cin>>n;
		cout<<"Enter No. Of Elements s =";
		cin>>s;
		cout<<"Enter Matrix Data\n";
		for(k=0;k<n;k++)
		{
			for(l=0;l<s;l++)
			{
				cin>>g[k][l];
			}
		}
		cout<<" Matrix Data    m[k][l]=(g[k][l]-0.5)/n;\n";
		for(k=0;k<n;k++)
		{
			for(l=0;l<s;l++)
			{
				m[k][l]=(g[k][l]-0.5)/n;
				cout<<m[k][l]<<" ";
			}
```

```
            cout<<endl;
        }
        sum=0.0;
        for(k=0;k<n;k++)
        {
            p=1.0;
            for(l=0;l<s;l++)
            {
            p=p*(1+(1.0/2.0*(m[k][l]-0.5))-(1.0/2.0*pow(m[k][l]-0.5,2)));
            }
            sum=sum+p;
        }
        cout<<"\nsum of middle value="<<sum;
        double res=pow(13.0/12.0,s);
        cout<<"\nresult of first part="<<res;
        res=res-(2.0/n*sum);
        cout<<"\nresult of first two part="<<res;
        getch();
         double y=0.0;
        double m1;
        for(k=0;k<n;k++)
        {
            double x=0.0;
            for(l=0;l<n;l++)
            {
                p=1.0;
                for(i=0;i<s;i++)
                {
m1=((1.0/2.0)*(m[k][i]-0.5))+((1.0/2.0)*(m[l][i]-0.5))-((1.0/2.0)*(m[k][i]-m[l][i]));
                    p=p*(1.0+m1);
                }
                x=x+p;
            }
            y=y+x;
```

```
        }
        cout<<"\nresult  y="<<y;
        double res1=res+(y/(n*n));
        cout<<"\nresult total="<<res1;
        cout<<"\nresult total as per square root="<<sqrt(res1);
        cout<<"\nCheck for another(Y/N)=";
        cin>>ch;
    }
}
```

Output

```
Enter No. Of Elements n=5
Enter No. Of Elements s =5
Enter Matrix Data
1 4 3 4 2
4 3 4 2 1
3 4 2 1 4
4 2 1 4 3
2 1 4 3 4
 Matrix Data    m[k][l]=(g[k][l]-0.5)/n;
0.1 0.7 0.5 0.7 0.3
0.7 0.5 0.7 0.3 0.1
0.5 0.7 0.3 0.1 0.7
0.7 0.3 0.1 0.7 0.5
0.3 0.1 0.7 0.5 0.7

sum of middle value=3.695155
result of first part=1.492143
result of first two part=0.014081
result  y=17.28
result total=0.705281
result total as per square root=0.83981
Check for another(Y/N)=
```

// program to find the Centered L_2 discrepancy with first row input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
	char ch='y';
	double p,sum,m[60][60];
	long k,l,s,i,n,g[60][60],j;
	while(ch=='y' || ch=='Y')
	{
		clrscr();
		cout<<"Enter No. Of Elements=";
		cin>>n;
		s=n;
		cout<<"\nEnter 1st Row Matrix Data\n";
		for(l=0,k=0;l<n;l++)
		{
			cin>>g[k][l];
		}
		for(k=1;k<n;k++)
		{
			for(l=0;l<n-k;l++)
			{
				g[k][l]=g[0][l+k];
			}
			for(j=0;l<n;l++,j++)
			{
				g[k][l]=g[0][j];
			}
		}
		clrscr();
```

```
cout<<" Matrix Data\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                cout<<g[k][l]<<" ";
        }
        cout<<endl;
}
cout<<" Matrix Data    m[k][l]=(g[k][l]-0.5)/n;\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                m[k][l]=(g[k][l]-0.5)/n;
                cout<<m[k][l]<<" ";
        }
        cout<<endl;
}
sum=0.0;
for(k=0;k<n;k++)
{
        p=1.0;
        for(l=0;l<n;l++)
        {
        p=p*(1+(1.0/2.0*(m[k][l]-0.5))-(1.0/2.0*pow(m[k][l]-0.5,2)));
        }
        sum=sum+p;
}
cout<<"\nsum of middle value="<<sum;
double res=pow(13.0/12.0,s);
cout<<"\nresult of first part="<<res;
res=res-((2.0/n)*sum);
cout<<"\nresult of first two part="<<res;
```

```
        double y=0.0;
        double m1;
        for(k=0;k<n;k++)
        {
            double x=0.0;
            for(l=0;l<n;l++)
            {
                p=1.0;
                for(i=0;i<n;i++)
                {
m1=((1.0/2.0)*(m[k][i]-0.5))+((1.0/2.0)*(m[l][i]-0.5))-((1.0/2.0)*(m[k][i]-m[l][i]));
                    p=p*(1.0+m1);
                }
                x=x+p;
            }
            y=y+x;
        }
        cout<<"\nresult  y="<<y;
        double res1=res+(y/(n*n));
        cout<<"\nresult total="<<res1;
        cout<<"\nresult total as per square root="<<sqrt(res1);
        cout<<"\nCheck for another(Y/N)=";
        cin>>ch;
    }
}
```

Output

```
 Matrix Data
1 4 3 4 2
4 3 4 2 1
3 4 2 1 4
4 2 1 4 3
2 1 4 3 4
 Matrix Data    m[k][l]=(g[k][l]-0.5)/n;
0.1 0.7 0.5 0.7 0.3
0.7 0.5 0.7 0.3 0.1
0.5 0.7 0.3 0.1 0.7
0.7 0.3 0.1 0.7 0.5
0.3 0.1 0.7 0.5 0.7

sum of middle value=3.695155
result of first part=1.492143
result of first two part=0.014081
result  y=17.28
result total=0.705281
result total as per square root=0.83981
Check for another(Y/N)=
```

//Program to find the Wrap-around discrepancy with first row input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
	 char ch='y';
	double p,sum,m[60][60],g[60][60],w[20];
	long k,l,s,i,n,j;
	while(ch=='y' || ch=='Y')
	{
		clrscr();
		cout<<"Enter No. Of Elements n=";
		cin>>n;
		s=n;
		cout<<"\nEnter 1st Row Matrix Data\n";
		for(l=0,k=0;l<n;l++)
		{
			cin>>g[k][l];
		}
		for(k=1;k<n;k++)
		{
			for(l=0;l<n-k;l++)
			{
				g[k][l]=g[0][l+k];
			}
			for(j=0;l<n;l++,j++)
			{
				g[k][l]=g[0][j];
			}
		}
		clrscr();
```

```
cout<<" Matrix Data\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                cout<<g[k][l]<<" ";
        }
        cout<<endl;
}
cout<<" Matrix Data    m[k][l]=(g[k][l]-0.5)/n;\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                m[k][l]=(g[k][l]-0.5)/n;
                cout<<m[k][l]<<" ";
        }
        cout<<endl;
}
for(k=1,j=0;k<=n;k++)
{
        for(l=k;l<=n-1;l++)
        {
                w[j]=l;
                j++;
        }
        w[j]=3;
        j++;
        for(l=1;l<=k-1;l++)
        {
                w[j]=l;
                j++;
        }
}
```

```
i=0;
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                g[k][l]=w[i];
                i++;
        }
}
cout<<" Matrix Data\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                cout<<g[k][l]<<" ";
        }
        cout<<endl;
}
 getch();
double res=pow(4.0/3.0,s);
cout<<"\nresult of first part="<<res;
double y=0.0;
double m1;
for(k=0;k<n;k++)
{
        double x=0.0;
        for(l=0;l<n;l++)
        {
                p=1.0;
                for(i=0;i<s;i++)
                {
                        m1=(m[k][i]-m[l][i])*(1.0-(m[k][i]-m[l][i]));
                         p=p*((3.0/2.0)-m1);
                }
```

```
                    x=x+p;
                }
                y=y+x;
            }
            cout<<"\nresult  y="<<y;
            double res1=res+(y/(n*n));
            cout<<"\nresult total="<<res1;
            cout<<"\nresult total as per square root="<<sqrt(res1);
            cout<<"\nCheck for another(Y/N)=";
            cin>>ch;
        }
}
```

Output

```
Enter No. Of Elements n=6

Enter 1st Row Matrix Data
1 2 3 4 5 5
 Matrix Data
1 2 3 4 5 5
2 3 4 5 5 1
3 4 5 5 1 2
4 5 5 1 2 3
5 5 1 2 3 4
5 1 2 3 4 5
 Matrix Data    m[k][l]=(g[k][l]-0.5)/n;
0.083333 0.25 0.416667 0.583333 0.75 0.75
0.25 0.416667 0.583333 0.75 0.75 0.083333
0.416667 0.583333 0.75 0.75 0.083333 0.25
0.583333 0.75 0.75 0.083333 0.25 0.416667
0.75 0.75 0.083333 0.25 0.416667 0.583333
0.75 0.083333 0.25 0.416667 0.583333 0.75

result of first part=5.618656
result  y=581.196612
result total=21.763006
result total as per square root=4.665084
Check for another(Y/N)=_
```

```
//Program to find the Wrap-around discrepancy matrix input
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
	char ch='y';
	double p,sum,m[60][60],g[60][60],w[20];
	long k,l,s,i,n,j;
	while(ch=='y' || ch=='Y')
	{
		clrscr();
		cout<<"Enter No. Of Elements n=";
		cin>>n;
		s=n;
		cout<<"\n Matrix Data\n";
		for(k=1,j=0;k<=n;k++)
		{
			for(l=k;l<=n-1;l++)
			{
				w[j]=l;
				j++;
			}
			w[j]=3;
			j++;
			for(l=1;l<=k-1;l++)
			{
				w[j]=l;
				j++;
			}
		}
		i=0;
```

```
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                g[k][l]=w[i];
                i++;
        }
}
cout<<" Matrix Data\n";
for(k=0;k<n;k++)
{
        for(l=0;l<n;l++)
        {
                cout<<g[k][l]<<" ";
        }
        cout<<endl;
}
getch();
cout<<" Matrix Data\n";
for(k=0;k<n;k++)
{
        for(l=0;l<s;l++)
        {
                m[k][l]=(g[k][l]-0.5)/n;
                cout<<m[k][l]<<"  ";
        }
        cout<<endl;
}
double res=pow(4.0/3.0,s);
cout<<"\nresult of first part="<<res;
double y=0.0;
double m1;
for(k=0;k<n;k++)
{
```

```
                double x=0.0;
                for(l=0;l<n;l++)
                {
                        p=1.0;
                        for(i=0;i<s;i++)
                        {
                                m1=(m[k][i]-m[l][i])*(1.0-(m[k][i]-m[l][i]));
                                p=p*((3.0/2.0)-m1);
                        }
                        x=x+p;
                }
                y=y+x;
        }
        cout<<"\nresult  y="<<y;
        double res1=res+(y/(n*n));
        cout<<"\nresult total="<<res1;
        cout<<"\nresult total as per square root="<<sqrt(res1);
        cout<<"\nCheck for another(Y/N)=";
        cin>>ch;
    }
}
```

Output

```
Enter No. Of Elements n=5
 Matrix Data
1 2 3 4 3
2 3 4 3 1
3 4 3 1 2
4 3 1 2 3
1 2 3 4 3
 Matrix Data
0.1  0.3  0.5  0.7  0.5
0.3  0.5  0.7  0.5  0.1
0.5  0.7  0.5  0.1  0.3
0.7  0.5  0.1  0.3  0.5
0.1  0.3  0.5  0.7  0.5

result of first part=4.213992
result  y=227.935563
result total=13.331414
result total as per square root=3.651221
Check for another(Y/N)=_
```

Chapter 5

// Program to find the Centered L_2-discrepancy with matrix input

```
#include<conio.h>
#include<stdlib.h>
#include<iostream.h>
#include<math.h>
#include<stdio.h>
void main()
{
	char ch='y';
	double p,sum,m[60][60];
	long k,l,s,i,n,j;
	while(ch=='y' || ch=='Y')
	{
		cout<<"Enter No. Of Elements n=";
		cin>>n;
		cout<<"Enter No. Of Elements s =";
		cin>>s;
		cout<<"Enter Matrix Data\n";
		for(k=0;k<n;k++)
		{
			for(l=0;l<s;l++)
			{
				cin>>m[k][l];
			}
		}
		sum=0.0;
		for(k=0;k<n;k++)
		{
			p=1.0;
			for(l=0;l<s;l++)
			{
			p=p*(1+(1.0/2.0*(m[k][l]-0.5))-(1.0/2.0*pow(m[k][l]-0.5,2)));
			}
```

```
                sum=sum+p;
        }
        cout<<"\nsum of middle value="<<sum;
        double res=pow(13.0/12.0,s);
        cout<<"\nresult of first part="<<res;
        res=res-((2.0/n)*sum);
        cout<<"\nresult of first two part="<<res;
        getch();
        double y=0.0;
        double m1;
        for(k=0;k<n;k++)
        {
                double x=0.0;
                for(l=0;l<n;l++)
                {
                        p=1.0;

                        for(i=0;i<s;i++)
                        {
m1=((1.0/2.0)*(m[k][i]-0.5))+((1.0/2.0)*(m[l][i]-0.5))-((1.0/2.0)*(m[k][i]-m[l][i]));
                                p=p*(1.0+m1);
                        }
                        x=x+p;
                }
                y=y+x;
        }
        cout<<"\nresult  y="<<y;
        double res1=res+(y/(n*n));
        cout<<"\nresult total="<<res1;
        cout<<"\nresult total as per square root="<<sqrt(res1);
        cout<<"\nCheck for another(Y/N)=";
        cin>>ch;
    }
}
```

Output

```
Enter No. Of Elements n=6
Enter No. Of Elements s =4
Enter Matrix Data
0.5632 0.2184 0.0546 0.1638
0.1645 0.2962 0.1348 0.4045
0.3700 0.0844 0.5001 0.0455
0.2531 0.1001 0.2695 0.3773
0.0914 0.6463 0.1967 0.0656
0.0914 0.2146 0.4048 0.2891

sum of middle value=3.02757
result of first part=1.377363
result of first two part=0.368173
result  y=12.926628
result total=0.727246
result total as per square root=0.852787
Check for another(Y/N)=_
```

LIST OF RESEARCH PAPERS FROM THE THESIS

Papers published / Communicated in peer refereed journals

1. Husain, B. and **Sharma, S.** (2015) - Optimal orthogonal designs in two blocks based on F-squares for mixture inverse model in four components. *International Journal of Experimental Design and Process Optimisation*, Vol. **4**, Nos. 3/4 pp. 206-215.

2. Husain, B. and **Sharma, S.** (2016) - Uniform Designs based on F – Squares. Accepted for publication in *International Journal of Experimental Designs and Process Optimisation.*

3. Husain, B. and **Sharma, S.** (2016) - Optimal Designs in two blocks based on F – Squares for reduced cubic canonical model in four components. *Communicated*.

4. Husain, B. and **Sharma, S.** (2016) - F-squares based efficient uniform designs for mixture experiments in four components. *Communicated*.

BIBLIOGRAPHY

1. Addleman, S. (1967). Equal and proportional frequency squares. *Journal of American Statistical Association*, **62**, 226-240.
2. Aggarwal, M. L., Sarin, V. and Singh, P. (2002) Optimal orthogonal block designs in two blocks for Becker's mixture models in three and four components. *Statistics and Probability Letters*, **59 (**4), 385–396.
3. Aggarwal M. L., Singh P. and Gupta N. (2004). Orthogonal block designs in two blocks for second degree K-model. *Statistics and Probability Letters*, **66**, 423-434.
4. Aggarwal M. L. and Singh P. (2006). D-optimal orthogonal designs in two orthogonal blocks for Darroch and Waller's quadratic model in constrained mixture components. METRON - *International Journal of Statistics*, **64** (3), 315-326.
5. Aggarwal M. L. and Singh P. (2008). Efficient uniform designs for mixture experiments designs in three and four components. *Trends in Applied Statistics Research, Nova Science Publications*, 11-26.
6. Aggarwal, M. L., Singh, P., Sarin, V., and Husain, B. (2008) Optimal orthogonal designs in two blocks based on F-Squares for Darroch and Waller's quadratic mixture model in four components. *Statistics and Applications*, **6** (1 & 2) (New Series), 242–259.
7. Aggarwal, M. L., Sarin, V., Singh, P. and Husain, B. (2009). Mixture Designs in orthogonal blocks using F-squares. METRON - *International Journal of Statistics*, LXVII (2), 105-128.
8. Aggarwal, M. L., Singh, P., Sarin, V. and Husain, B. (2011a). Nearly optimal orthogonally blocked designs for four mixture components based on F–squares. *Communications in Statistics – Simulation and Computation,* **40 (**1), 177–195.
9. Aggarwal, M. L., Singh, P., Sarin, V. and Husain, B. (2011b). Orthogonally blocked mixture component-amount designs via projection of F–squares. *Journal of the Korean Statistical Society,* **41 (**1), 49–60.
10. Aggarwal, M. L., Singh, P., Sarin, V. and Husain, B. (2013). Optimal orthogonal block designs for four mixture components in two blocks based on F–squares for Becker's and K-model. *Statistics: A Journal of theoretical and Applied Statistics*, **47** (5), 1003–1021.

11. Aitchison, J. and Bacon-Shone, J. (1984). Log contrast models for experiments with mixtures. *Biometrika*, **71**, 323-330.

12. Becker, N. G. (1968). Models for the response of a mixture. *Journal of the Royal Statistical Society*, Series B, **30**, 349-358.

13. Box, G. E. P. and Hau, I. (2001). Experimental designs with one or more factor constraints. *Journal of Applied Statistics*, **28** (8), 937-989.

14. Box, G. E. P. and Hunter, J. S. (1957). Multi-factor experimental designs for exploring response surfaces. *Annals of Mathematical Statistics*, **28**, 195-242.

15. Bundschuh, P. and Zhu, Y. C. (1993). A method for exact calculation of the discrepancy of low-dimensional finite point sets (I). *Abhandlungen aus dem Math. Seminar der Univ. Hamburg*, Bd., 63.

16. Chan, L. Y. (2000). Optimal designs for experiments with mixtures: a survey. *Communications in Statistics, Theory and Methods*, **29**, 2281-2312.

17. Chan, L. Y., Gaun, Y. N. and Zhang, C. Q. (1998a). A-optimal designs for additive quadratic mixture model. *Statistica Sinica*, **8**, 979-990.

18. Chan, L. Y., Meng, J. H., Jiang, Y. C. and Gaun, Y. N. (1998b). D-optimal axial designs for quadratic and cubic additive mixture models. *Australian and New Zealand Journal of Statistics,* **40**, 359-371.

19. Chan, L. Y. and Sandhu, M. K. (1999) Optimal orthogonal block designs for a quadratic mixture model for three components. *Journal of Applied Statistics*, **26** (1), 19–34.

20. Chantarat, N., Allen, T.T., Ferhatosmanoglu, N. and Bernshteyn, M. (2006). A combined array approach to minimize expected prediction errors in experimentation involving mixture and process variables**.** *International Journal of Industrial and Systems Engineering*, **1**, 129-147.

21. Chung, P. J., Goldfarb, H. B., and Montgomery, D. C. (2007). Optimal Designs for Mixture-Process Experiments with Control and Noise Variables. *Journal of Quality Technology*, **39**, 179–190.

22. Clarke, G. M. (1968). Inverse polynomial response surfaces applied to data from plant nutrition experiments. *Biometrics*, **24**, 226-227.

23. Clarke, G. M. and Esan, E. O. (1971). Inverse polynomial response surfaces applied to data from plant nutrition experiments. *Biometrics*, **27**, 246.

24. Cornell, J. A. (1973). Experiments with mixtures: a review. *Technometrics*, **15**, 437-455.

25. Cornell, J. A. (1975). Some comments on designs for Cox's mixture polynomial. *Technometrics*, **17**, 25-35.
26. Cornell, J. A. (1988). Analyzing data from mixture experiments containing process variables: a split-plot approach. *Journal of Quality Technology*, **20**, 2-23.
27. Cornell, J. A. (1995). Fitting models to data from mixture experiments containing other factors. *Journal of Quality Technology*, **27**, 13-33.
28. Cornell, J. A. (2000). Fitting a stack-variable model to mixture data: some questions raised. *Journal of Quality Technology*, **32**, 133-147.
29. Cornell, J. A. (2002). Experiments with mixtures. 3rd ed. *Wiley, New York*.
30. Cornell, J. A. and Gorman, J. W. (1984). Fractional design plans for process variables in mixture experiments. *Journal of Quality Technology*, **16**, 20-38.
31. Cox, D. R. (1971). A note on polynomial response functions for mixtures. *Biometrika*, **58**, 155-159.
32. Crosier, R. B. (1984). Mixture experiments: geometry and pseudo-components. *Technometrics*, **26**, 209-216.
33. Crosier, R. B. (1986). The geometry of constrained mixture experiments. *Technometrics*, **28**, 95-102.
34. Czitrom, V. (1988). Mixture experiments with process variables: D-optimal orthogonal experimental designs. *Communications in Statistics*, *Theory and Methods*, **17** (1), 105–121.
35. Czitrom, V. (1989). Experimental design for four components with process variables. *Communications in Statistics*, *Theory and Methods*, **18**, No. 12, 4561–4581.
36. Czitrom, V. (1992). Note on a mixture experiment with process variables. *Communications in Statistics Part B: Simulation and Computation*, **21**, 493–498.
37. Daniel, C. (1963). Discussion on Professor Scheffė's paper. *Journal of the Royal Statistical Society*, *B*, **25**, 256-257.
38. Darroch, J. N. and Waller, J. (1985). Additivity and interactions in three component experiments with mixtures. *Biometrika*, **72**, No. 1, 153–163.
39. Draper, N. R and John, R. C. S. (1977). A mixture model with inverse terms. *Technometrics*, **19** (1), 37–46.

40. Draper, N. R., Prescott, P., Lewis, S. M., Dean, A. M., John, P. W.M. and Tuck, M. G. (1993). Mixture designs for four components in orthogonal blocks. *Technometrics*, **35** (3), 268–276.

41. Draper, N. R and Pukelshiem, F. (1997). Mixture models based on homogeneous polynomials. *Journal of Statistical Planning and Inference*, **71**, 303-311.

42. Draper, N. R and Pukelshiem, F. (1999). Kiefer ordering of simplex designs for first- and second-degree mixture models. *Journal of Statistical Planning and Inference*, **79**, 325-348.

43. Dueck, G. and Scheuer, T. (1990). Threshold accepting: a general purpose algorithm appearing superior to simulated annealing. *J. Comput. Phys.,* **90**, 161-75.

44. Fang, K. T. (1980). The uniform design: application of number-theoretic methods in experimental design. *Act a Math. Appl. Sinica*, **3**, 363-372.

45. Fang, K. T. and Wang, Y. (1981). A note on uniform distribution and experimental design. *KeXue TongBao*, **26**, 485-489.

46. Fang, K. T. and Wang, Y. (1994). Number – Theoretic Methods in Statistics. Chapman & Hall, London.

47. Fang, K. T. and Li, J. K. (1995). Some new results on uniform design. *Chinese Sci. Bull*, **40**, 268-272.

48. Fang, K. T., Lin, D. K. J., Winker, P. and Zhang, Y. (2000). Uniform designs: Theory and applications. *Technometrics*, **42**, 237-248.

49. Fang, K. T. and Winker, P. (1998). Uniformity and orthogonality. *Technical Report MATH*-175, Hong Kong Baptist University, Dept. of Mathematics.

50. Fang, K. T., Ma, C. X. and Winker, P. (2001). Centered L_2-discrepancy of random sampling and Latin hypercube design and construction of uniform design. *Math. Comput.*, **71**, 275-296.

51. Fang, K. T. and Mukherjee, R. (2000). A connection between uniformity and aberration in regular functions of two-level factorials. *Biometrika*, **87**, 193-198.

52. Fang, K. T., Ma, C. X. and Winker, P. (2001). Centered L_2-discrepancy of random sampling and Latin hypercube design and construction of uniform design. *Math. Comput.* **71**, 275-296.

53. Fang, K. T., Shiu, W. C. and Pan, J. X. (1999). Uniform designs based on Latin squares. *Statistica Sinica*, **9**, 905-912.

54. Fang, K. T. and Yang, Z. H. (2000). On uniform design of experiments with restricted mixtures and generation of uniform distribution on some domains. *Statistics & Probability Letters*, **46**, 113-120.

55. Finney, D. J. (1945). Some orthogonal properties of 4×4 and 6×6 Latin squares. *Annals of Eugenics*, **12**, 213-219.

56. Finney, D. J. (1946a). Orthogonal partitions of 5×5 Latin Squares. *Annals of Eugenics* **13**, 1-3.

57. Finney, D. J. (1946b)**.** Orthogonal partitions of 6×6 Latin Squares. *Annals of Eugenics* **13**, 184-196.

58. Freeman, G. H. (1966). Some non-orthogonal partition of 4×4, 5×5 and 6×6 Latin Squares. *Annals of Mathematical Statistics*, **37**, 661-681.

59. Ghosh, S. and Liu, T. (1999). Optimal mixture designs for four components in orthogonal blocks, *J. Statist. Plan. Inference*, **78**, 219–228.

60. Goos, P. and Donev, A. N. (2006). The D-optimal design of blocked experiments with mixture components. *Journal of Quality Technology*, **38** (4), 319–332.

61. Gorman, J. W. (1970). Fitting equations to mixture data with restraints on compositions. *Journal of Quality Technology*, **2**, 186-194.

62. Gorman, J. W. and Cornell, J. A. (1982). A note on a model reduction for experiments with both mixture components and process variables. *Technometrics*, **24**, 243-247.

63. Gorman, J. W. and Hinman, J. E. (1962). Simplex-lattice designs for multicomponent systems. *Technometrics*, **21**, 159-173.

64. Hare, L. B. (1974). Mixture designs applied to food formulation. *Food Technology*, **28**, 50-62.

65. Hare, L. B. (1979). Designs for mixture experiments involving process variables. *Technometrics*, **4**, 463-487.

66. Hedayat, A. and Seiden, E. (1970). F-squares and orthogonal F-squares design: A generalization of latin square and orthogonal latin square design. *Annals of Mathematical Statistics*, **41**, 2035-2044.

67. Hedayat, A., Raghavarao, D. and Seiden, E. (1975). Further contributions to the theory of F-squares design. *Annals of Mathematical Statistics*, **3**, 712-716.

68. Hilgers, R.D. and Heiligers, B. (2003). A note on optimal mixture and mixture amount designs. *Statistica Sinica*. **13**, 709-725.

69. Hilgers, R. D. (2000). D-optimal design for Becker's minimum polynomial. *Statistics and Probability Letters*, **49**, 175-179.

70. Hickernell, F. J. (1998a). A Generalized Discrepancy and Quadrature Error Bound. *Mathematics of Computation,* **67***,* 299-322.

71. Hickernell, F. J. (1998b). Lattice rules: how well do they measure up? *In Random and Quasi-Random point Sets*, 106-166. Springer-Verlag, New York.

72. Hickernell, F. J., Liu, M. Q. and Yam, C. Y. (2000). Discrepancy measure of uniformity. *Journal Chinese Statistics Association*, **38**, 353-373.

73. Hua, L. K. (1956). Introduction to Number Theory. *Science Press, Beijing.*

74. Hua, L. K. and Wang, Y. (1981). Applications of Number Theory to Numerical Analysis. *Springer and Science Press, Berlin and Beijing.*

75. John, P. W. M. (1984). Experiments with mixture involving process variables, 1–17, Technical Report **8**, *Center for Statistical Sciences, University of Texas*, TX.

76. Kiefer, J. C. (1959). Optimum experimental designs. *J. Roy. Statist. Soc. Ser. B* **21**, 272–304.

77. Kiefer, J. C. (1975). Optimal design: variation in structure and performance under change of criterion. *Biometrika*, **62**, 277–288.

78. Kiefer, J. C. (1978). Asymptotic approach to families of design problems. *Comm. Statist. Theory Methods*, A **7**, 1347–1362.

79. Klein, T. (2004). Optimal designs for second-degree kronecker model mixture experiments. *Journal of Statistical Planning and Inference*, **123**, 117-131.

80. Kowalski, S., Cornell, J. A. and Vining, G. G. (2000). A new model and class of designs for mixture experiments with process variables. *Communications in Statistics, Theory and Methods*, **29**, 312-341.

81. Kurotori, I. S. (1966). Experiments with mixtures of components having lower bounds. *Industrial Quality Control*, **22**, 592-596.

82. Laywine, C. F. (1989). A geometric construction for sets of MOFS. *Utilas Mathemetica*, **35**, 95-102.

83. Lee, H. H. and Warner, J. C. (1935). The system biphenyl-bibenzyl-naphthalene: nearly ideal binary and ternary systems. *Journal of the American Chemical Society*, **37**, 318-321.

84. Lewis, S. M., Dean, A. M., Draper, N. R. and Prescott, P. (1994) Mixture designs for q components in orthogonal blocks. *Journal of the Royal Statistical Society*, Series B, **56**, 457-467.

85. Ma, C. X. (1997a). A new criterion of uniformity – symmetrical discrepancy. *Journal of Nankai University,* **30**, 30-37.

86. Ma, C. X. (1997b). Construction of uniform designs using symmetrical discrepancy. *Applied Statistics Management*, **16**, 166-169.

87. McKay, M. D., Beckman, R. J. and Conover, W. J. (1979). A comparison of three methods for selecting values of input variables in the analysis of output from a computer code. *Technometrics,* **21**, 239-245.

88. Mclean, R. A. and Anderson, V. L. (1966). Extreme vertices design of mixture experiments. *Technometrics,* **8**, 447-454.

89. Murty, J. S. and Das, M. N. (1968). Design and analysis of experiments with mixtures. *Annals of Mathematical Statistics*, **39**, 1517-1539.

90. Myers, R. H. and Montgomery, D. C. (2002). Response Surface Methodology. 2^{nd} *edn. John Wiley & Sons, Inc., New York.*

91. Naes, T., Faergestad, E. M. and Cornell, J. A. (1998). A comparison of methods for analyzing data from a three-component mixture experiment in the presence of variation created by two process variables. *Chemometrics & Intelligent Laboratory Systems*, **41**, 221-235.

92. Nelder, J. A. (1966). Inverse polynomial response, a useful group multifactor response function. *Biometrics*, **22**, 128-141.

93. Niederreiter, H. (1978b). Quasi-Monte Carlo Methods and Pseudo-random Numbers. *Bull. Amer. Math. Soc.*, **84**, 957-1041.

94. Niederreiter, H. and Peart, P. (1986). Localization of search in quasi-Monte Carlo methods for global optimization, *SIAM J. Sci. Statist. Computing,* **7**, 660-664.

95. Niederreiter, H. (1988a). Quasi-Monte Carlo methods for multi-dimensional numerical integration. *International Series of Numerical Mathematics,* Birk. Springer-Verlag.

96. Niederreiter, H. (1992). Random number generation and Quasi-Monte Carlo Methods. *SIAM CBMS-NSF Regional Conference Series in Applied Mathematics. Philadelphia.*

97. Nigam, A. K. (1970). Block designs for mixture experiments, *Annals of Mathematical Statistics*, **41** (6), 1861–1869.

98. Nigam, A. K. (1976). Corrections to blocking conditions for mixture experiments. *Annals of Mathematical Statistics*, **47** (6), 1294–1295.

99. Nigam, A. K., Gupta, S. C. and Gupta, S. (1983). A new algorithm for extreme vertices designs for linear mixture models. *Technometrics*, **25**, 367-371.

100. Piepel, G. F. (1983a). Defining consistent constraints regions in mixture experiments. *Technometrics*, **25**, 97-101.

101. Piepel, G. F. (1983b). Calculating centroids in constrained mixture experiments. *Technometrics*, **25**, 279-283.

102. Piepel, G. F. and Cornell, J. A. (1985). Models of mixture experiments when the response depends on the total amount. *Technometrics*, **27**, 219-227.

103. Piepel, G. F. and Cornell, J. A. (1987). Designs for mixture-amount experiments. *Journal of Quality Technology*, **19**, 11-28.

104. Piepel, G. F. and Cornell, J. A. (1994). Mixture experiment approaches: examples, discussion and recommendations. *Journal of Quality Technology*, **26**, 177-196.

105. Piepel, G. F. and Cornell, J. A. (2008). A catalog of mixture experimental examples. *BS-SA*-3298, Rev. 16.0, *Battelle, Pacific Northwest Division Richland, Washington*.

106. Prescott, P., Draper, N. R., Dean, A. M. and Lewis, S. M. (1993). Mixture designs for five components in orthogonal blocks, *Journal of Applied Statistics*, **20** (1), 268–276

107. Prescott, P., Draper, N. R., Lewis, S. M. and Dean, A. M. (1997). Further properties of mixture designs for five components in orthogonal blocks. *Journal of Applied Statistics*, **24**, 147-156.

108. Prescott, P. and Draper, N. R. (1998). Mixture designs for constrained components in orthogonal blocks. *Journal of Applied Statistics*, **25**, 613-638.

109. Prescott, P. (2000). Projection designs for mixture experiments in orthogonal blocks. *Communications in Statistics – Theory and Methods*, **29** (9-10), 2229-2253.

110. Prescott, P., Draper, N.R., Dean, A.M. and Lewis, S.M. (2002). Mixture experiments: ILL-conditioning and quadratic model specification. *Technometrics*, **44** (3), 260-268.

111. Saxena, S. K. and Nigam, A. K. (1973). Symmetric-Simplex Block Designs for mixtures, *Journal of the Royal Statistical Society.* Series B *(Methodological),* **35** (3), 466-472.

112. Saxena, S. K. and Nigam, A. K. (1977). Restricted exploration of mixtures by symmetric-simplex designs. *Technometrics* **19**, 47-52.

113. Scheffé, H. (1958). Experiments with mixtures, *Journal of the Royal Statistical Society*, Series B, **20** (2), 344–360.

114. Scheffé, H. (1963). Simplex centroid designs for experiments with mixtures, *Journal of the Royal Statistical Society*, Series B, **25** (2), 235–263.

115. Shaw, J. E. H. (1988). A quasi random approach to integration in Bayesian Statistics. *Ann. Statist.*, **16**, 895-914.

116. Singh, P. (2003). Optimal orthogonal designs in two blocks for Darroch and Waller's quadratic mixture model in three and four components, *METRON-International Journal of Statistics*, **61** (3), 419–430.

117. Smith, W. F. and Beverly, T. A. (1997). Generating linear and quadratic Cox mixture models. *Journal of Quality Technology*, **29**, 211-224.

118. Snee, R. D. (1973). Techniques for the analysis of mixture data. *Technometrics*, **15**, 517-528.

119. Snee, R. D. (1975). Experimental designs for quadratic models in mixture spaces. *Technometrics*, **17** (2), 149-159.

120. Snee, R. D. (1979). Experimental designs for Mixture System with Multicomponent constraints. *Communications in Statistics – Theory and Methods*, **8**, 303-326.

121. Snee, R. D. and Marquardt, D. W. (1974). Extreme vertices design for linear mixture models. *Technometrics*, **16**, 399-408.

122. Snee, R. D. and Marquardt, D. W. (1976). Screening concepts and designs for experiments with mixtures. *Technometrics*, **18**, 19-29.

123. Thompson, W. O. and Myers, R. H. (1968). Response surface designs for experiments with mixtures. *Technometrics*, **10**, 739-756.

124. Vuchkov, I. N., Damgaliev, D. L. and Yontchev, C. A. (1981). Sequentially generated second-order quasi D-optimal designs for experiments with mixture and process variables. *Technometrics*, **23**, 233-238.

125. Wang, Y. and Fang, K. T. (1990). Number – theoretic Methods in Applied Statistics. *Chinese Annals of Mathematics*, Series B, **11**, 859-914.

126. Wang, Y. and Fang, K. T. (1996). Uniform design of experiments with mixtures. *Science in China (Ser. A)*, **39**, 264-275.

127. Warnock, T. T. (1972). Computational investigations of low discrepancy point sets. In *Applications of Number Theory to Numerical Analysis* (Edited by S. K. Zaremba), 319-343. Academic Press, New York.

128. Wynn, H. P. (1970). The Sequential generation of D-optimal experimental designs. *Annals of Mathematical Statistics*, **41**, 1655-1664.

Chapter 3

1. The expressions of the eigenvalues for the Reduced Cubic Canonical Model for Design 1, Design 2 and Design 3, case a = 0 and c = 0 for Optimal Block Design.

2. The expressions of the eigenvalues for the Reduced Cubic Canonical Model for Design 1, Design 2 and Design 3 for Optimal Orthogonal Block Design.